Dielectric Properties of SBR Vulcanizates Loaded with HAF Carbon Black and $BaTiO_3$ Ceramics

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ABSTRACT: The influence of HAF carbon black and $BaTiO_3$ ceramic powder contents in SBR vulcanizates on the dielectric constant (ε') at different frequencies and at fixed temperature of 303 K is studied well in this article. The temperature dependence of the ac conductivity (σ_{ac}) was also studied. ε' appreciably decreases as frequency increased for both filled and unfilled SBR vulcanizates. At each frequency, ε' gradually decreased with $BaTiO_3$ loading, but its change at any fixed frequency with $BaTiO_3$ filler loading is not uniform. For HAF group ε' (at loading $\geq$ 40 phr), drops rapidly with frequency. Meanwhile, it increased appreciably beyond a certain HAF filler loading ($\approx$ 20 phr). Experimental values of the dielectric constant of both $BaTiO_3$ and HAF contents were compared with those calculated by using Tsangaris, Clausius and Bruggman models. Tsangaris model with simple modifications was applied and a fairly good agreement was obtained. The HAF particles or aggregates was found to take the shape of oblate ellipsoids with the minor axes parallel to the applied frequency as detected from the decreasing behavior of the depolarizing factor (Y) with HAF contents. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 103: 2227–2234, 2007

Key words: SBR; HAF; $BaTiO_3$; dielectric; ac conductivity

INTRODUCTION

Composite consisting of a polymer matrix and dispersed ceramic particles is a kind of materials with great potential properties and applications. By integrating the advantages of the two phases, the composite materials can offer enhanced performances far beyond those of the individual constituent materials.[1,2] Generally, polymers are flexible and processable, but their dielectric constant and piezoelectric coefficient are relatively low, while ceramics have high dielectric constant and piezoelectric coefficient.[3] For these reasons, the application of an individual polymer or a kind of ceramic is greatly restricted in many aspects, the preparation of ceramic/polymer composite materials by dispersing ceramic particles into polymer matrix provides a new route in combining the merits of polymers and ceramics. Varieties of ceramic/polymer composites have been investigated to enhance certain performances, including the dielectric properties. Possessing very high dielectric constant, barium titanate ($BaTiO_3$) based ceramic has been widely utilized as capacitors and piezoelectric transducers.[4–6]

On the other hand, dielectric measurements are generally available techniques to measure the properties of polymer composites that are adequate for electronic applications. It is also very useful to characterize the polymeric materials which are marked with polar groups in their structures. The orientation of such polarizable entities under electric field can involve molecular chain and create dielectric relaxation phenomena in polymer.[7] In addition, the study of dielectric relaxation provides information about the dielectric polarization and relaxation. The former is related to the magnitude of the electric moment and the latter is essentially a function of the molecular dimensions. These two dielectric parameters are not necessarily related to each other. In general, studying of the dielectric properties of the dielectric materials is useful for understanding of the insulating properties and the structure for each material.

Practical polymeric systems are almost heterogeneous. The conductive or non- conductive fillers are usually not mixed but dispersed in the polymer matrix.[8] The effect of such noncompatible additives to the dielectric permittivity of composites has been studied using a great variety of formulas used to calculate the dielectric permittivity, ε, of the composite as a function of its components volume fractions, V_i, and permittivities, ε_i. These formulas can be represented in general form.[9]

$$\varphi(\varepsilon) = \sum_{i=1}^{n} v_i \varphi(\varepsilon_i) \qquad (1)$$

i.e., not the values of ε and ε_i themselves but some functions φ of these magnitudes obey the simple

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arithmetic law of mixing. Different relationships have been also proposed for $\varphi(\varepsilon)$.[9–12] The dielectric constant of mixed dielectrics can also be calculated by means of special formulas of mixtures.[13–20] Most of these formulas proposed for the permittivity of two phase systems have two limitations.[21] First, the dispersed particles are either assumed to be spherical or their shape is not explicitly included in the equations. Second, the dielectric permittivity is assumed to be independent of the applied field frequency. However the most remarkable effect developed in heterogeneous dielectrics is the interfacial polarization or Maxwell-Wagner Sillars (MWS) effect. This polarization occurs as a result of the accumulation of virtual charges at the interface of two media, having different permittivites and/or conductivities.[22]

As a continuation of a previous work,[23] the present article aims to discuss the effect of $BaTiO_3$ powder ceramic and HAF carbon black contents embedded in SBR matrix on the dielectric constant and the ac conductivity. The experimental results are checked using theoretical models.

EXPERIMENTAL

The properties of the rubber composites depend largely on the preparation method,[24] such as the order of adding the ingredients, time of mixing and processing. So it is very important that all samples must have the same circumstances and pass throughout the same procedure. The rubber composites were prepared by using a two roll mill of 300 mm length, 170 mm diameter with speed of slow roll 18 rev/min and gear ratio 1.4.

The prepared compounded rubber was left for at least 24 h before vulcanization. The vulcanization process was conducted at 140°C under a pressure of 40 kg/cm^2 for 30 min. For reasonable stability and reproducibility of parameter, samples were subjected to thermal aging at 343 K for 25 days in an electrical oven[25] before measurements were made.

The test materials have the compositions shown in Tables I and II. They were prepared from commercial ingredients according to standard techniques[26]

TABLE I
Composition of SBR Samples Containing Different Concentrations of HAF Carbon Black

Ingredient (phr)	Samples			
SBR(1502)	100	100	100	100
HAF(N-330)	0	20	40	60
Processing oil	10	10	10	10
Stearic acid	2	2	2	2
MBTS	2	2	2	2
PBN	1	1	1	1
Zinc oxide	5	5	5	5
Sulfur	2	2	2	2

TABLE II
Composition of SBR Samples Containing Different Concentrations of $BaTiO_3$

Ingredient (phr)[a]	Samples				
SBR(1502)	100	100	100	100	100
HAF(N-330)	40	40	40	40	40
$BaTio_3$	0	10	30	60	100
Processing oil	10	10	10	10	10
Stearic acid	2	2	2	2	2
MBTS[b]	2	2	2	2	2
PBN[c]	1	1	1	1	1
Zinc oxide	5	5	5	5	5
Sulfur	2	2	2	2	2

[a] Part per hundred parts of rubber by weight.
[b] Dibenzthiazyl disulphide.
[c] Pheny1-β-naphthylamine.

by using the facilities of the Transport and Engineering (Rubber Manufacturing) Company (Trenco), Alexandria, Egypt.

The sample then is coated with silver past and sandwiched between two parallel plates of brass electrodes which were isolated from each other using Teflon. The capacitance was measured at different temperatures and frequencies using an LCZ meter type (Keithley 3321-1).

The dielectric constant ε' (real part of the dielectric constant) of the samples was calculated by using the relation

$$\varepsilon' = \frac{d}{\varepsilon_0 A} C \qquad (2)$$

where C is the capacitance of the sample, d is the thickness of the sample, A is the cross-sectional area of each of the parallel surfaces of the sample and ε_0 is the permittivity of free space $= 8.85 \times 10^{-12}$ F/m.

RESULTS AND DISCUSSIONS

Frequency dependence

Plots of the dielectric constant (ε') as a function of frequency at constant temperature of 303 K for different $BaTiO_3$ loadings (in 40 HAF/SBR samples) are presented in Figure 1. The dispersion region spreads over the whole frequency range (i.e., 10^2–10^5 Hz) used for measurement. The dielectric constant appreciably decreases as frequency increased for both filled and unfilled systems. The nature of variation of the dielectric constant with frequency for $BaTiO_3$ filled systems is very much similar to that of the unfilled one, but the extent of dispersion is lower for filled systems. At each frequency, the dielectric constant gradually decreases with $BaTiO_3$ loading, but the rate of change of dielectric constant at any fixed frequency with $BaTiO_3$ filler loading is not uniform. The dielectric constant ε' for the case of HAF black

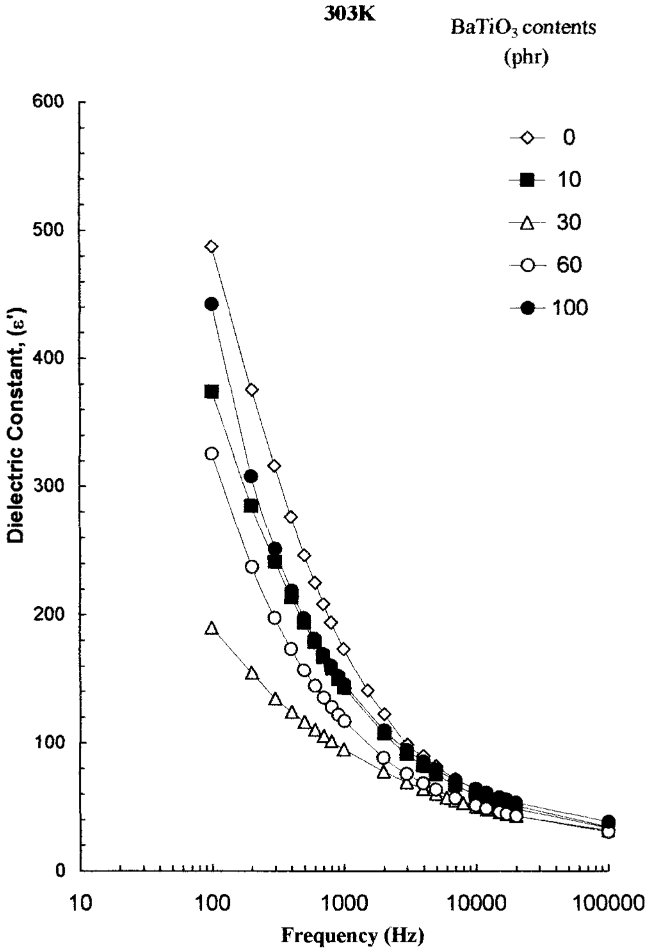

Figure 1 The frequency dependence of dielectric constant for (40HAF/SBR) composites having different $BaTiO_3$ contents at 303 K.

loaded rubber samples (≥ 40 phr), drops rapidly with frequency and becomes relatively smaller at higher frequency 10^5 Hz as shown in Figure 2. Meanwhile ε′ changes slightly with frequency for the low loaded rubber samples.

Filler dependence

Unexpectedly, the dielectric constant at 100 Hz (cf. Fig. 3) of all mixes loaded with concentrations < 30 phr $BaTiO_3$ decreased due to addition of $BaTiO_3$ in spite of the polar nature of the $BaTiO_3$. This appears the masking role of HAF black inside the SBR matrix. For relatively high $BaTiO_3$ loadings (> 30 phr loading), the dielectric constant of SBR sample increases.

Meanwhile, the dielectric constant, ε′ increases appreciably beyond a certain HAF filler loading (≅ 20 phr) as shown in Figure 4. In this region a relatively small increase in filler loading produces a large increase in the dielectric constant. Further increase in HAF loading beyond the critical concentration region causes abrupt change in the dielectric constant as well as the ac conductivity (σ_{ac}) (as shown in Fig. 5) of SBR composites. Thus, for HAF filled SBR composites there is a marginal change in ε′ and ac conductivity for (10–20) phr loading variation and then ε′ and σ_{ac} show a sudden increase, giving rise to a very sharp transition from a low to a very high conductive material for just 40 phr of HAF black loading. Meanwhile, $BaTiO_3$ filler loading does not contribute very well to both ε′ and σ_{ac} for SBR loaded with 40 phr HAF black (cf. Figs. 3 and 6). This difference is attributed to the fact that the inherent aggregation form of HAF black has a higher tendency to form a three-dimensional network in the composites ensuring better electrical response than the $BaTiO_3$ filler.[27]

Theoretical models

The dielectric constant of the composite materials containing more than one component can be expressed in the general form as in eq. (1):

$$F(\varepsilon') = \sum_{i=1}^{m} v_i F(\varepsilon'_i) \tag{3}$$

where $F(\varepsilon')$ is some function of the composite dielectric constant, v_i and ε_i are volume fraction and dielectric

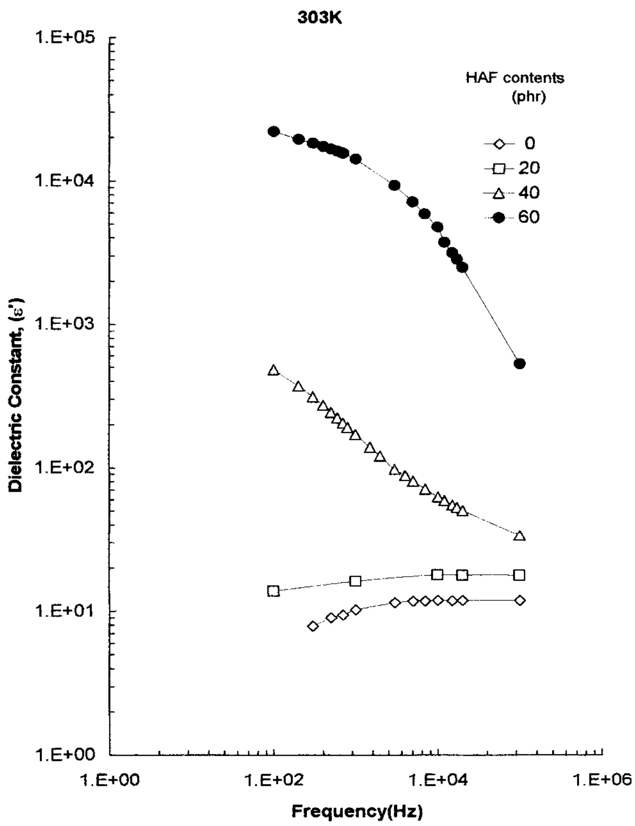

Figure 2 The frequency dependence of dielectric constant for different HAF contents of SBR composites at 303 K.

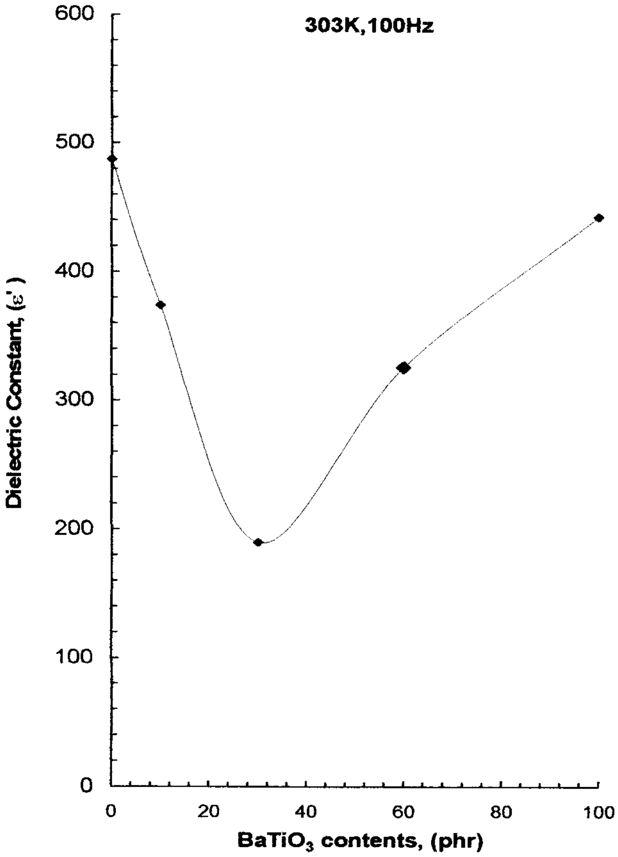

Figure 3 The dependence of dielectric constant on $BaTiO_3$ contents in (40HAF/SBR) composites at 303 K and 100 Hz.

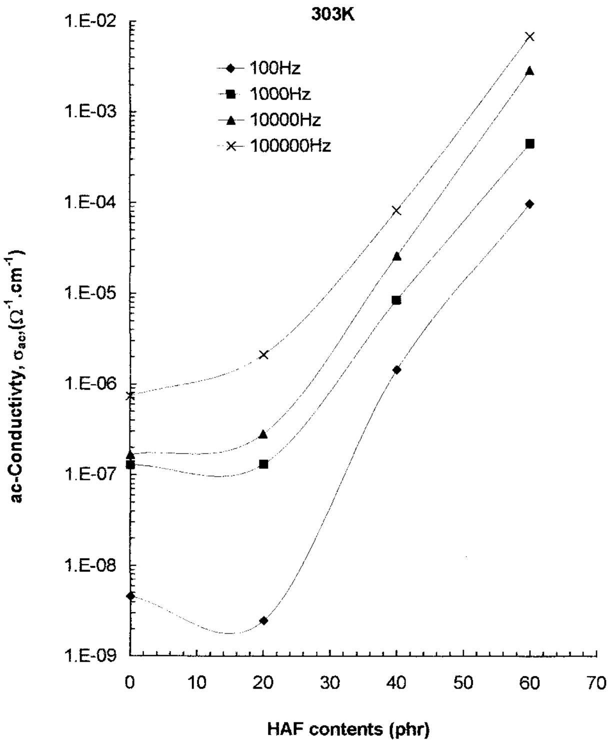

Figure 5 The dependence of ac-conductivity on HAF contents of SBR composites at 303 K for different frequencies.

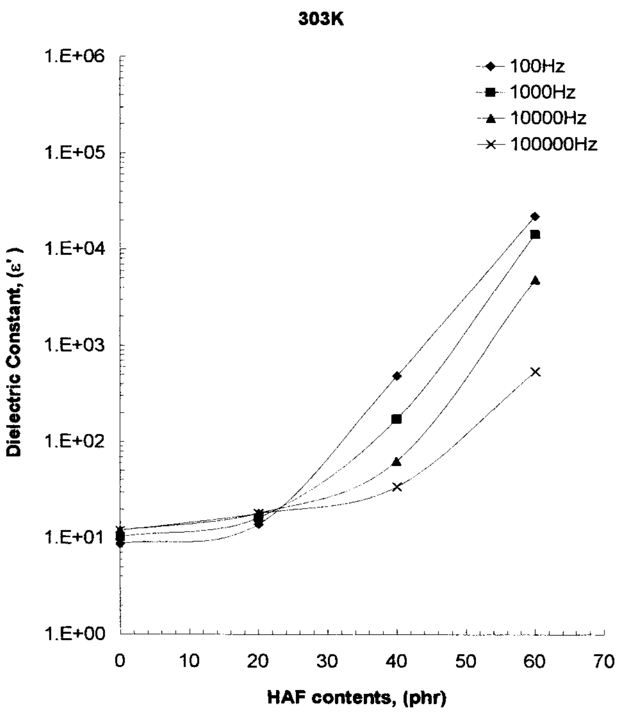

Figure 4 The dependence of dielectric constant on HAF contents of SBR composites at 303 K for different frequencies.

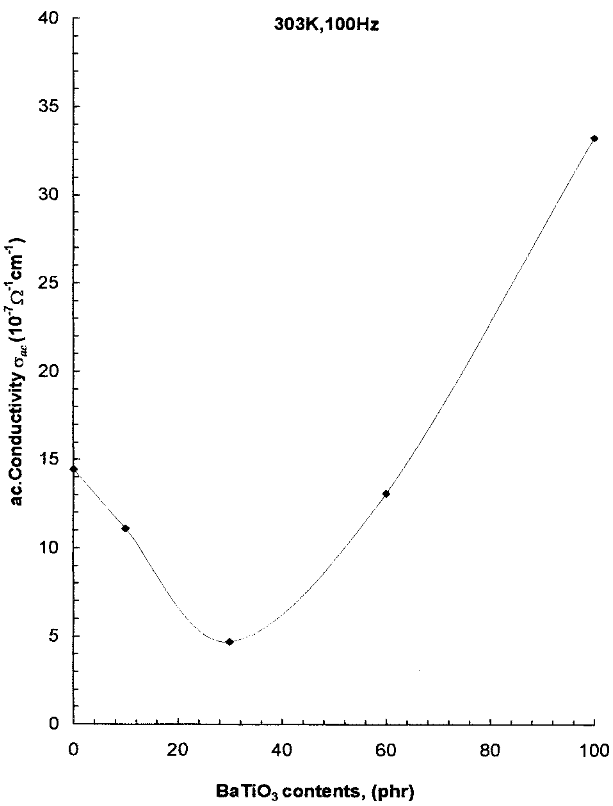

Figure 6 The dependence of ac conductivity on $BaTiO_3$ contents in (40HAF/SBR) composites at 303 K and 100 Hz.

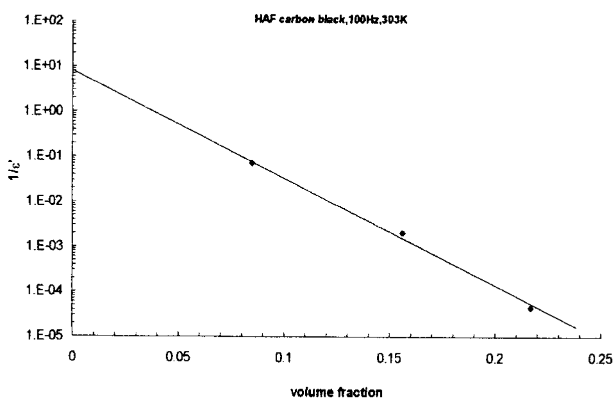

Figure 7 The dependence of $1/\varepsilon'$ on HAF volume fraction of SBR composites at 303 K and 100 Hz.

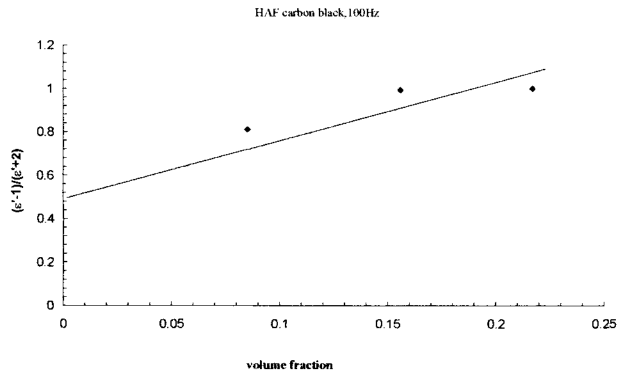

Figure 9 The dependence of $(\varepsilon' - 1)/(\varepsilon' + 2)$ on HAF volume fraction of SBR composites at 303 K and 100 Hz

constant for the *i*th component of the composite containing a number of *m* components.

For a two-component system, eq. (3) assumes the form:

$$F(\varepsilon') = v_1\varepsilon'_1 + \varepsilon'_2(1 - v_1) \tag{4}$$

where ε'_1 and ε'_2 are the dielectric constants of components 1 and 2, and v_1 is the volume fraction of component 1.

Figures 3 and 4 illustrate the dielectric interaction pattern of mixed systems, with respect to composition. The dielectric constants ε' in these plots are those observed at a frequency of 100 Hz. The plot of ε' versus v_2, the volume fraction of $BaTiO_3$ and carbon black (HAF) systems shows clear nonlinear. However, plots of $1/\varepsilon'$ against v_2 (cf. Figs. 7 and 8) which describes the law of harmonic mixture, are linear for HAF carbon black and $BaTiO_3$.

This linearity of plots reflects the series combination of the constituent dielectrics in the composites. Extrapolation of the plots to $v_2 = 0$ yields $\varepsilon' = \varepsilon'_1$, where ε'_1 the dielectric constant of the vulcanized gum SBR matrix at $f = 100$ Hz. Graphically obtained ε'_1 value is in disagreement with the experimentally obtained value, which gives for the case of HAF ε'_1 Graphically is 0.1 and experimental is 8.6 and for the $BaTiO_3$ loading 125.57 and 487.25 respectively.

A trial was made to test the applicability of the Clausius-Mossotti equation for a mixture of dielectrics in the present systems. Linear plots are obtained when specific polarization $[(\varepsilon' - 1)/(\varepsilon' + 2)]$ is plotted against v_2 for both systems (HAF/SBR and $BaTiO_3$/SBR) (cf. Figs. 9 and 10). Slopes and intercepts obtained are different for HAF black and $BaTiO_3$-rubber systems. Dielectric constant for vulcanized SBR rubber can be theoretically obtained from the intercepts of these plots at $v_2 = 0$. Graphical values for ε'_1 for two systems are 4.46 and 150 respectively, where corresponding experimental values are found to be 8.6 and 487.25, respectively.

The calculated values of dielectric constant at 100 Hz for $BaTiO_3$ rubber system are shown in Table IV. The observed values of ε' are lower than the theoretical values from existing relationship of heterogeneous dielectrics at the same frequency, according to the Clausius-Mossotti equation:

$$\varepsilon' = \frac{(1 - v_2)2\varepsilon_2'^2 + (1 + 2v_2)\varepsilon'_1\varepsilon'_2}{(1 - v_2)\varepsilon'_1 + (2 + v_2)\varepsilon'_2} \tag{5}$$

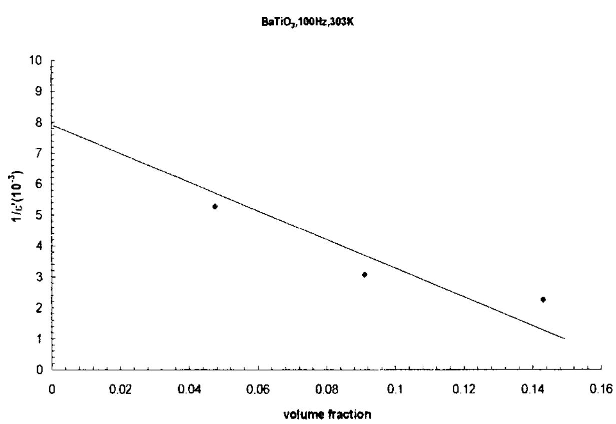

Figure 8 The dependence of $1/\varepsilon'$ on $BaTiO_3$ volume fraction of 40HAF/SBR composites at 303 K and 100 Hz.

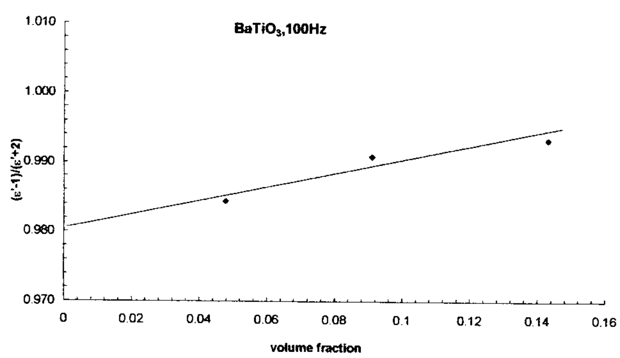

Figure 10 The dependence of $(\varepsilon' - 1)/(\varepsilon' + 2)$ on $BaTiO_3$ volume fraction of SBR composites at 303 K and 100 Hz.

One of the successful models is that of Bruggman (Ref. 14 cited in Ref. 28), which assumes:

$$\left(\frac{\varepsilon - \varepsilon_1}{\varepsilon_1 - \varepsilon_2}\right)\frac{\varepsilon_1}{\varepsilon} = (1 - v_2)^3 \tag{6}$$

He considered the case of spherical conductive inclusions and the last equation is simplified by assuming that the permittivity of the conductive inclusions tends to infinity. It is then transformed to eq. (7), which is often used, provided that the volume fraction of the inclusions is considerably less than unity ($v_2 \ll 1$).

$$\varepsilon' = \varepsilon'_1(I + 3v_2) \tag{7}$$

Subscript (1) refers to the matrix and subscript (2) to the filler (for SBR samples only contains HAF black), but for SBR mixes contains constant concentration HAF (40 phr) and different concentration of $BaTiO_3$, ε' in eq. (7) is considered as the matrix dielectric constant and ε'_1 in eq. (7) is the dielectric constant of 40 HAF/SBR matrix. The calculated values of ε' for both SBR mixes are given in Tables III and IV.

The deviation between observed values and theoretical values [calculated using eq. (7)] for HAF/SBR system at higher HAF loading indicates that the shape, size, and distribution of HAF particles do not permit a high degree of interaction in the present system.[17] Meanwhile, there is a great disagreement in both values and behavior of ε' in $BaTiO_3$/SBR system.

In all the above formulae the dielectric constant is supposed to be independent of both the applied frequency and the components characteristics. This was taken care of by Tsangaris et al.,[29] who proposed a new model with suitable equations formulated to expressing dielectric constant ε' and dielectric loss ε'' of composite materials as a function of the applied frequency and the component characteristics. These equations are as follows:

$$\varepsilon' = \frac{\varepsilon'_1}{[(\varepsilon'_1 - 1)^Y + 1]}\left\{\left[\left(\frac{\sigma}{\omega\varepsilon_0}\right)^{v_2}(\varepsilon'_1 - 1)^{1-v_2}\cos\frac{\pi v_2}{2}\right]^Y + 1\right\} \tag{8}$$

TABLE III
Dielectric Constant Obtained from Theoretical Models (Bruggman and Tsangaris) and Experimental Values for HAF Black/SBR Composites

HAF volume fraction	ε' Bruggman (100 Hz)	Tsangaris (100 Hz)	Experimental (100 Hz)
0	8.69	8.69	8.69
0.085	10.9	18.207	13.92
0.156	12.76	469.97	487.25
0.217	14.35	21791	22271

TABLE IV
Dielectric Constant Obtained from Theoretical Models (Clausius, Bruggman, and Tsangaris) and Experimental Values for $BaTiO_3$/SBR Composites

$BaTiO_3$ volume fraction	ε' Clausius (100 Hz)	Bruggman (100 Hz)	Tsangaris (100 Hz)	Experimental (100 Hz)
0	487.25	11.74	490.82	495.8
0.016	497.148	12.32	360	364.367
0.048	516.37	13.48	167.24	166.445

where Y is the depolarizing factor given by[30]

$$Y = \frac{1}{1 - (a/b)^2} - \frac{a/b}{\left[1 - (a/b)^2\right]^{1/2}}\cos^{-1} a/b \tag{9}$$

where ω is the frequency, σ is the conductivity of the filler, ε_o is the dielectric constant of the free space, ε'_o the dielectric constant of the matrix, and *a/b* is the aspect ratio of the filler (*a* and *b* are the principal axes).

The application of eq. (8) which gives the dielectric constant of (HAF/SBR) composites (at constant frequency $\cong$100 Hz)) as a function of HAF volume fractions is shown together with experimental values in Figure 11 (at room temperature (303 K)). Figure 11 shows an increasing behavior of dielectric constant of the SBR as the HAF filler volume fraction increase as predicted by theory[31,32] and found by many workers.[33–36] The experimental ε' values are in a good agreement with calculated one [as shown in Table III].

As the difference between the conductivities of the host polymer [(SBR) and/or ($BaTiO_3$/SBR composites)] and HAF filler is extremely great, very strong MWS effect[37] is expected because of the incompatibility between rubber (soft phase) and carbon black (hard phase)in such systems. This phenomenon appears in heterogeneous media consisting of phases with different dielectric constant and conductivities and is due to accumulation of charges at interfaces.

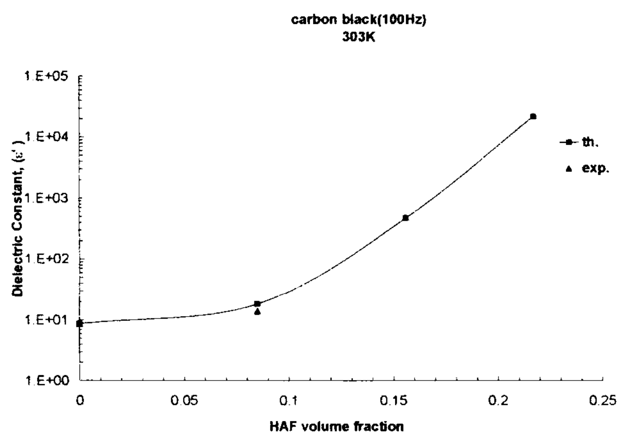

Figure 11 Dielectric constant of (HAF/SBR) composites as a function of HAF volume fractions at 303 K and 100 Hz.

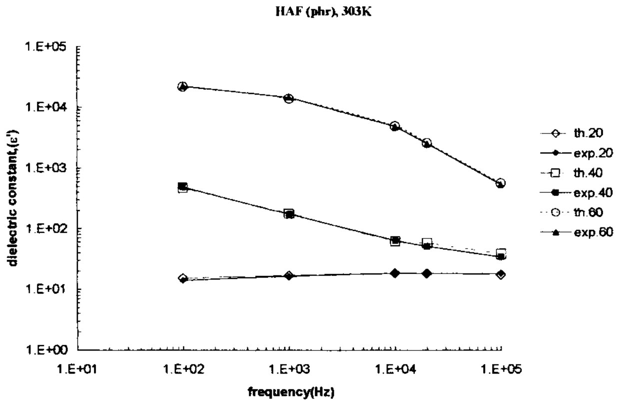

Figure 12 The frequency dependence of dielectric constant of SBR composites having different HAF black contents at 303 K.

The proposed model approaches the experimental values more closely, for a low as well as for high volume fraction of HAF conductive material. Moreover, Figure 12 shows the dielectric constant of different contents of HAF black in SBR samples as a function of frequency. It shows the expected decrease in dielectric constant with increasing frequency, which occurs since the interfacial polarization which dominates in the low frequency region gradually diminishes to zero at high frequencies.[33,38]

For the case of $BaTiO_3$ filler in SBR samples the proposed model of Tsangaris et al. is also checked with ε_1' (the dielectric constant of SBR mixes with $BaTiO_3$ only) calculated from equation (3.16). A fairly good agreement between the modified proposed models with the experimental values for all $BaTiO_3$ volume fractions is found as shown in Figure 13 and Table IV.

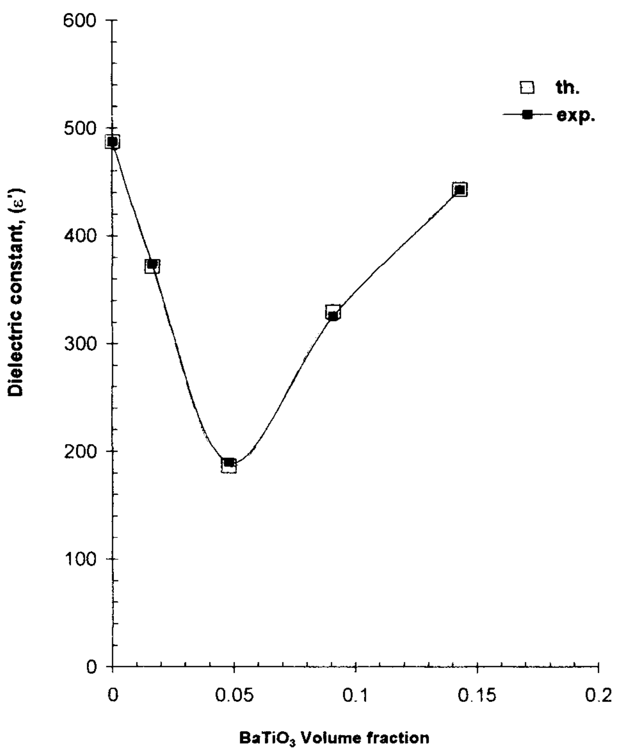

Figure 13 Dielectric constant of ($BaTiO_3$/SBR) composites as a function of $BaTiO_3$ volume fractions at 303 K and 100 Hz.

The shape of HAF black may be transformed from spherical to ellipsoidal or even to a long rod shape according to the volume fraction of HAF within the matrix and presence of another filler (such as $BaTiO_3$) as well as both temperature and frequency (which is not taken into consideration by Tsangaris et al.). The aspect ratio *a/b* of HAF black and thus the depolarizing factor (Y) are assumed to vary with the variation of the above-mentioned parameters. Values of the depolarizing factor (Y), which were chosen to fit the calculated dielectric constant with the experimental ones, are ranged from (0.33–1.7) for temperature and frequency.

The depolarizing factor (Y) for high frequencies ($> 10^4$ Hz) and for 40 phr of HAF/SBR loaded with

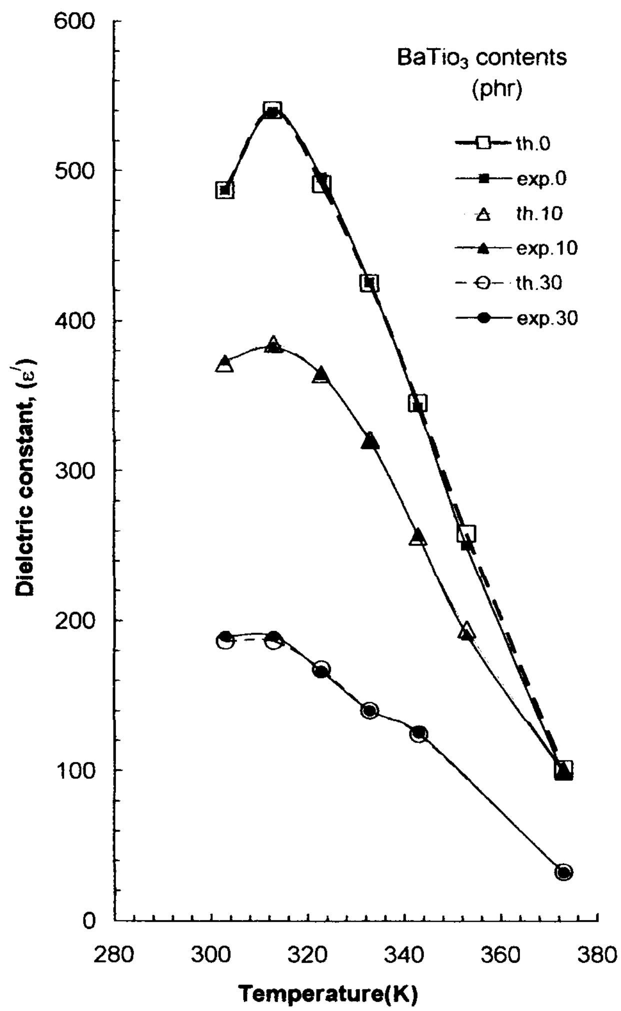

Figure 14 Temperature dependence of dielectric constant of SBR composites having different concentration of $BaTiO_3$ at 100 Hz.

30 phr $BaTiO_3$ sample is substituted with the value that is responsible for spherical inclusions with *a/b* ≅ 1, so $Y = 0.333$. The HAF particles or aggregates take the shape of oblate ellipsoids with the minor axes (*a*) parallel to the applied frequency.[39]

Temperature dependence

The temperature dependence of the dielectric constant for composites containing 40 phr of HAF carbon black and 0, 10, and 30 phr of $BaTiO_3$ is presented in Figure 14. When conductive particles are dispersed in a non conductive matrix, the dielectric constant of the composite increases with the volume fraction of the filler as predicted by theory and by this work (as seen in the last subsection). It can also be seen that composites contain 40 phr of HAF black with different $BaTiO_3$ contents (0–30 phr) exhibit a slight increase in ε' at low temperatures followed by a gradual decrease with increasing temperature as shown in Figure 14. It is clearly noticed that, differential thermal expansion of the SBR matrix and HAF black disrupts the chains of contacting particles and decreases the dielectric constant. Tsangaris et al. model also describes quite satisfactorily the decrease in the dielectric constant with increasing temperature (for 10–30 phr of $BaTiO_3$ contents) as clearly observed in Figure 14.

CONCLUSIONS

From the above discussions it is concluded that

1. The dielectric constant, ε', appreciably decrease as frequency is increased both for filled and unfilled SBR system.
2. At each frequency, ε', gradually decreases with $BaTiO_3$ loading.
3. Dielectric constant increases appreciably beyond a certain HAF loading (≈ 20 phr).
4. The experimental ε' values for both groups of samples are in good agreement with the calculated one based on Tsangaris model.
5. The depolarizing factor Y for high frequency and 40 phr of HAF/SBR loaded with 30 phr $BaTiO_3$ sample is substituted by a value which is responsible for spherical inclusion with *a/b* ≅ 1.
6. A slight increase followed by gradual decrease in, ε', with temperature was detected for samples loaded with different $BaTiO_3$ contents.

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