

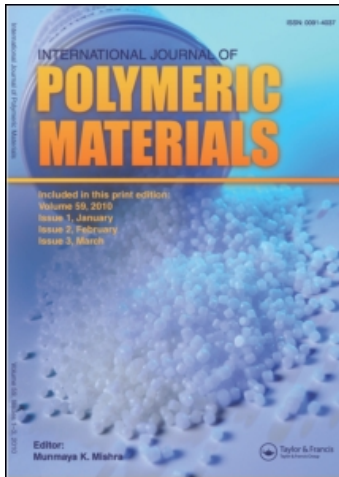
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### Mechanical and dielectric properties of styrene-butadiene rubber polyester short-fiber composites: Part II. Composites loaded with high abrasion furnace carbon black

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## **MECHANICAL AND DIELECTRIC PROPERTIES OF STYRENE-BUTADIENE RUBBER POLYESTER SHORT-FIBER COMPOSITES: PART II. COMPOSITES LOADED WITH HIGH ABRASION FURNACE CARBON BLACK**

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*The mechanical and dielectric properties of styrene butadiene rubber (SBR) reinforced with polyester (PE) short-fiber loaded with high abrasion furnace (HAF) carbon black in concentrations up to 60 phr have been investigated. The effect of ageing on these properties was also studied. It was found that the tensile strength and young's modulus increased with increasing carbon content in SBR–PE fiber composites, while elongation at break was decreased. These properties showed the same trend with ageing time, too.*

*The permittivity and the dielectric loss have been determined at a frequency range from 0.1 KHz to 10 MHz at temperatures between 30 and 60° C for the same samples. It was found that in samples having low carbon concentrations, both the permittivity and the dielectric loss slightly increased with the increase of carbon concentration whereas at high concentrations they showed more pronounced anomalous dispersion with frequency. The dielectric loss-frequency curves were broad, indicating a distribution of relaxation times. The data were analyzed using a computer program based on Fröhlich terms. The results of the analysis were discussed and interpreted.*

*Keywords:* styrene-butadiene rubber, polyester short-fiber, high abrasion furnace, dielectric properties, anomalous dispersion, relaxation time

## **INTRODUCTION**

Synthetic polymers have found extensive applications as electrical insulators. For extruded cables, they have wide applications ranging from power to control cables. A good combination of flexibility and strength is an essential requirement of cable insulator. A continuous effort is being made

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to improve the mechanical and thermal endurance properties of cable insulators without much sacrifice of the electrical properties.

Styrene butadiene rubber (SBR) is now extensively employed for insulation and sheathings where it has an economic advantage over natural rubber [1].

Short fibers attracted the attention of several researchers due to their good dispersion in and good adhesion to rubber matrix [2, 3] as well as their advantage on the physico-mechanical and dielectric properties. The adhesion between many types of short fibers and most elastomers have been achieved by the discovery of the tricomponent system HRH (Hydrated silica, Resorcinol, Hexamethylene tetramine) [4–7]. Compounding of rubber with short fibers have been studied before [8].

In a previous paper [9] the effect of addition of a tricomponent adhesion system HRH to styrene-butadiene rubber (SBR) polyester (PE) short-fiber composites, on the mechanical and dielectric properties was reported. It was found that the presence of HRH system in these composites improved the mechanical and dielectric properties and resist ageing. Also the effect of polyester short-fiber concentration in SBR vulcanizates was investigated. It was found that an increase of PE short fiber content in SBR improves the mechanical and dielectric properties. The effect on the mentioned properties of adding another reinforcing agent to SBR–PE fiber composites was studied [9]. Twenty phr (parts per hundred parts rubber) of SRF (Semi-Reinforcing Furnace) carbon black, whose particle size is 60 nm, were added to SBR–PE fiber vulcanizates. It was found that the mechanical and dielectric properties improved. Moreover, the presence of carbon black in these composites facilitated the compounding of SBR with PE short-fiber. Therefore more PE short-fiber could be added to SBR vulcanizates.

In the present paper we examine in greater detail the influence of the addition of another type of carbon black, HAF (High Abrasion Furnace) whose particle size is 28 nm, to SBR–PE fiber composite, with varying concentrations from zero up to 60 phr, on the mechanical and dielectric properties at different frequencies and temperatures. In addition, the effect of thermal and natural ageing of these composites on the mentioned properties has been investigated.

## EXPERIMENTAL

### Materials

- (1) Styrene-butadiene rubber (SBR) 1502 supplied by Esso Chemie.
- (2) Zinc oxide to activate the action of the accelerator.
- (3) *N*-cyclohexyl-*z*-benzothiazole sulfonamide (CBS) acts as an accelerator to reduce the time required for cure.
- (4) Stearic acid acts as a softener to facilitate the dispersion of material added to rubber.

- (5) Naphthenic processing oil (sp.gr. 0.96 and viscosity at 100°C = 80–90 CP).
- (6) Colloidal Hydrated silica (Hisil), Resorcinol and Hexamethylenetetramine (HMTA) as tricomponent adhesive system (HRH).
- (7) Sulfur (SP. Gr. 2.04–2.06) essential vulcanizing agent.
- (8) Polyester (PE) short-fiber [64mm] from Misr company silk, Kafr El-Dawar Egypt.
- (9) High Abrasion Furnace (HAF) carbon black N330.

### Sample Preparation

The preparation of rubber vulcanizates were carried out according to ASTM method [10]. All ingredients were accurately weighed. Mixing was carried out on a laboratory controlled temperature two roll mill of the following dimensions: outside diameter 470 mm, working distance = 300 mm, speed of slow roll = 17 r.p.m and friction ratio 1 : 1.4. Care was taken to ensure fiber orientation in the mill direction [11].

### Mechanical Measurements

The apparatus used for measuring the processing and curing characteristics of the rubber compounds have been described in the previous paper [9]. The measurements were carried out at  $150 \pm 1^\circ\text{C}$ . The vulcanized sheets were cut into five individual dumbbell-shaped specimens in longitudinal and transversal directions by steel die of constant width (0.4 cm) the minimum thickness of test specimens was determined by gauge calibrated to a hundredth of a millimeter.

Tensile strength, elongation at break and Young's modulus of the samples were determined as before [9].

### Accelerated Ageing

The oven method [12] was used where thermal ageing of rubber vulcanizates was carried out in an oven at  $90 \pm 1^\circ\text{C}$  for different periods of time. The mechanical properties were determined after ageing and compared with those before ageing.

The retained values of the samples under investigation were determined using the equation.

$$\text{Retained value} = \frac{\text{property after ageing}}{\text{property before ageing}} \times 100 \quad (1)$$

### Dielectric Measurements

Measurements of the permittivity  $\epsilon'$  and the dielectric loss  $\epsilon''$  for the samples under investigation were carried out as described in the previous paper [9].

The errors in  $\epsilon'$  and  $\epsilon''$  amounts to  $\pm 2\%$  and  $\pm 5\%$  respectively. The samples were prepared in the form of discs 50 mm in diameter and 3 mm thick.

## RESULTS AND DISCUSSION

To study the effect of adding HAF carbon black to SBR–PE fiber composites on the mechanical and dielectric properties, samples loaded with different concentrations up to 60 phr HAF black were prepared as given in Table 1.

### Mechanical Properties

The rheometric characteristics are given in Table 2. From Table 2 it is clear that the maximum torque ( $M_H$ ) and the optimum cure time ( $t_{C90}$ ) decrease with the initial loading of HAF carbon black, then increase with further increase of HAF black content and finally attain a constant value, while the reverse is true for the cure rate index CRI.

**TABLE 1** Rubber formulations containing different concentrations of HAF carbon black

Ingredients	Phr
SBR 1502	100.00
Stearic acid	2.00
Zinc oxide	5.00
Processing oil	3.00
Hydrated silica	5.00
Hexamethylene-Tetramine	3.20
Resorcinol	5.00
CBS	1.00
Sulfur	2.00
Polyester fiber	20.00
HAF carbon black	0–60

**TABLE 2** Rheometric characteristics at  $(152 \pm 1^\circ\text{C})$  for SBR–PE fiber mixes containing different HAF carbon black contents

Property	Sample	HAF carbon black contents (phr)							
		Control sample	0	10	20	30	40	50	60
$M_L$	(dNm)	60.00	77.00	60.90	62.50	65.60	72.50	71.00	71.50
$M_H$	(dNm)	8.50	7.00	3.90	4.90	6.00	6.80	6.40	6.50
$t_{S2}$	(min)	7.50	3.65	3.75	3.25	2.25	2.25	2.25	2.25
$t_{C90}$	(min)	18.50	24.00	19.25	22.50	22.50	23.00	23.50	23.50
CRI	$(\text{min})^{-1}$	9.10	4.90	6.45	5.20	5.01	4.82	4.82	4.82

The mechanical properties were measured for the rubber vulcanizates in the longitudinal and transversal directions. The data obtained are given in Table 3 and illustrated graphically in Figure 1. From this figure it is clear

**TABLE 3(a)** Longitudinal values of the tensile strength in (N/mm<sup>2</sup>) before and after thermal ageing of (SBR–PE)/HAF composites

Carbon black contents ( phr)	Before ageing	After ageing (days)				
		1	2	4	6	7
Control	3.37	3.39	3.31	3.26	3.16	2.94
0	4.95	5.19	5.23	5.33	5.39	5.73
10	7.96	8.35	8.37	8.57	8.84	9.27
20	10.52	11.21	11.39	11.84	11.98	12.18
30	12.65	13.54	13.87	14.32	14.97	15.18
40	15.82	16.98	17.40	17.99	18.95	19.21
50	18.36	19.71	20.40	20.99	22.00	22.65
60	19.95	21.65	22.21	22.99	23.76	24.79

**TABLE 3(b)** Transversal values of the tensile strength in (N/mm<sup>2</sup>) before and after thermal ageing of (SBR–PR)/HAF composites

Carbon black contents ( phr)	Before ageing	After ageing (days)				
		1	2	4	6	7
Control	3.88	3.86	3.53	3.21	3.00	2.96
0	4.45	4.55	4.56	4.59	4.63	4.68
10	6.24	6.39	6.47	6.65	6.89	6.95
20	8.75	8.95	9.11	9.43	9.67	9.79
30	10.36	10.53	10.65	10.70	10.98	11.11
40	12.95	13.22	13.51	13.64	13.83	13.87
50	13.45	14.16	14.28	14.45	14.65	14.78
60	15.36	16.42	16.45	16.51	16.89	16.99

**TABLE 3(c)** Longitudinal values of the elongation at break % before and after thermal ageing of (SBR–PE)/HAF composites

Carbon black contents ( phr)	Before ageing	After ageing (days)				
		1	2	4	6	7
Control	545	490	465	415	355	320
0	184	168	145	126	115	100
10	162	149	137	120	117	110
20	154	138	127	118	107	100
30	141	130	121	113	103	95
40	121	111	100	95	80	75
50	116	106	97	81	76	63
60	96	84	78	68	60	50

**TABLE 3(d)** Transversal values of the elongation at break % before and after thermal ageing of (SBR-PE)/HAF composites

Carbon black contents ( phr)	Before ageing	After ageing (days)				
		1	2	4	6	7
Control	550	510	475	418	366	325
0	200	180	170	156	141	124
10	185	164	146	126	106	96
20	177	145	136	112	85	77
30	162	129	103	85	73	63
40	143	101	85	73	64	54
50	133	86	72	53	42	36
60	118	81	61	42	30	26

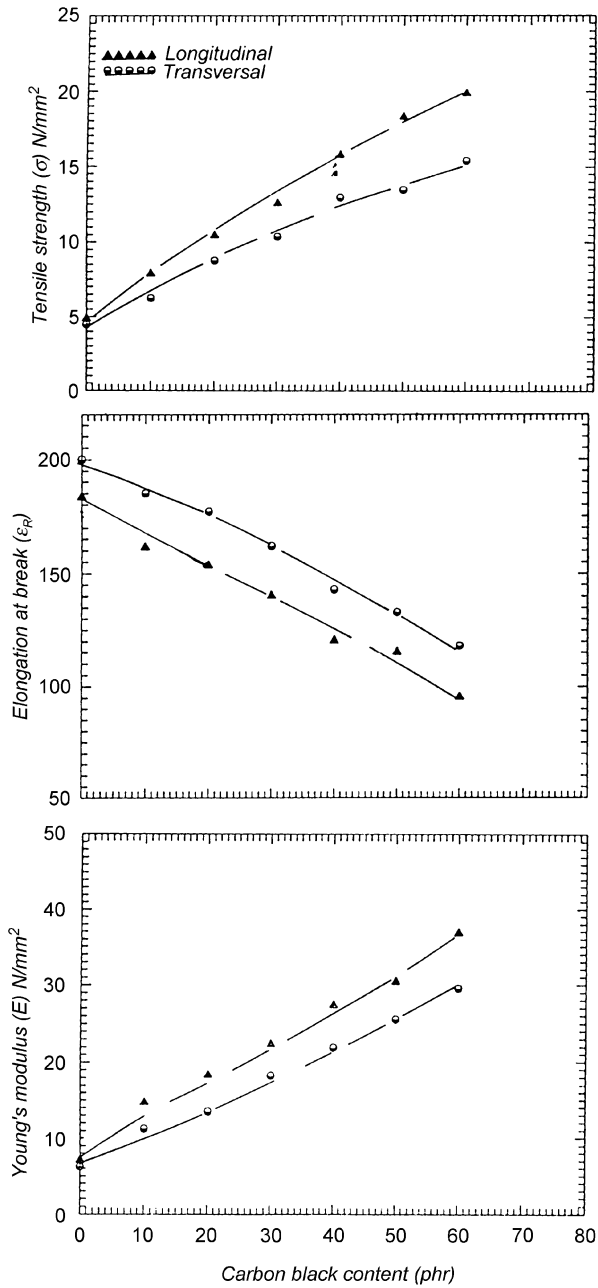
**TABLE 3(e)** Longitudinal values of young's modulus in (N/mm<sup>2</sup>) before and after thermal ageing of (SBR-PE)/HAF composites

Carbon black contents ( phr)	Before ageing	After ageing (days)				
		1	2	4	6	7
Control	1.89	1.92	1.94	1.96	2.00	2.05
0	7.23	7.57	7.69	8.45	9.36	9.57
10	14.85	17.25	19.07	19.85	20.25	22.95
20	18.50	21.00	22.26	23.90	26.00	28.30
30	22.50	24.60	25.50	27.90	29.66	32.60
40	27.50	29.00	30.90	32.50	33.00	34.56
50	30.60	31.77	32.50	33.27	34.60	35.00
60	37.00	37.90	38.82	39.67	40.53	41.96

**TABLE 3(f)** Transversal values of young's modulus in (N/mm<sup>2</sup>) before and after thermal ageing of (SBR-PE)/HAF composites

Carbon black contents ( phr)	Before ageing	After ageing (days)				
		1	2	4	6	7
Control	1.37	1.39	1.39	1.42	1.46	1.51
0	6.21	6.21	6.23	6.25	6.29	6.53
10	11.25	12.77	13.95	16.27	18.56	21.42
20	13.50	15.21	17.24	19.56	22.46	22.58
30	18.22	20.00	22.11	24.07	27.27	28.00
40	21.89	23.95	25.55	27.95	34.26	32.56
50	25.56	26.87	27.90	30.05	32.72	34.22
60	29.56	31.00	32.11	33.30	34.56	35.16

that the tensile strength and Young's modulus increase with the increase of HAF carbon content, while elongation at break decreases in both directions. Also it is obvious that values of the tensile strength and Young's modulus in



**FIGURE 1** Dependence of the mechanical properties (Long-Trans) for SBR-PE fiber vulcanizates on HAF black content.



the longitudinal direction are higher than those in the transversal one, while an opposite trend is observed for elongation at break, this indicates that the polyester short-fiber was fairly oriented in the longitudinal direction inside the rubber matrix.

## **Effect of Ageing on the Mechanical Properties**

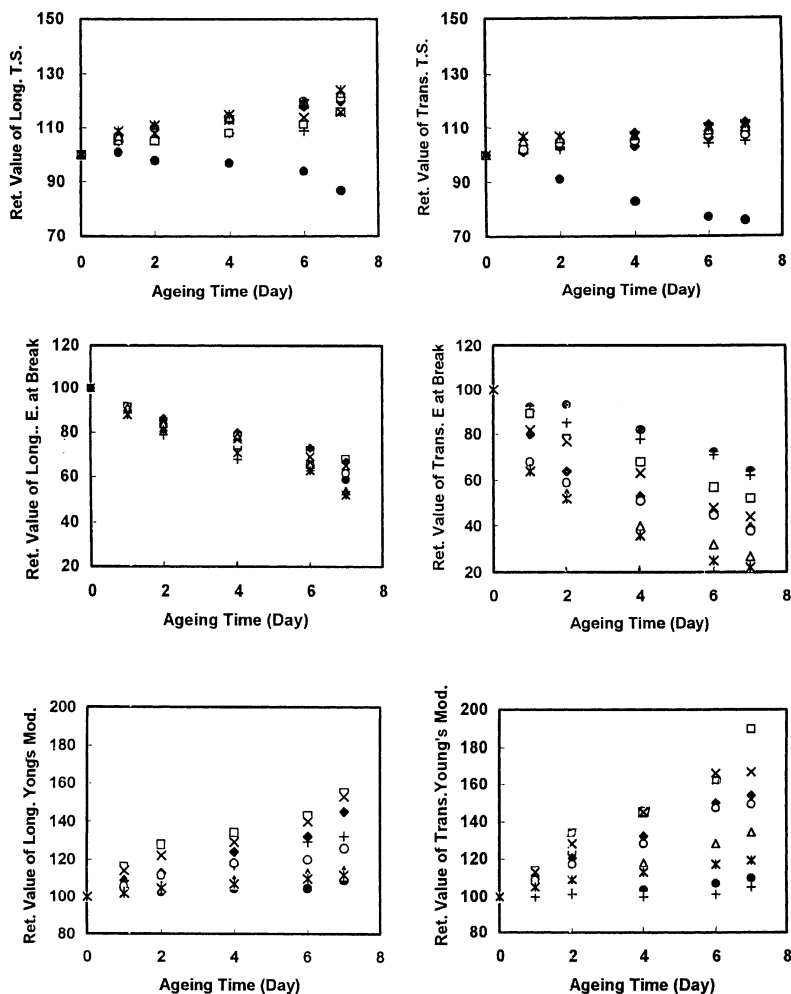
### ***Thermal Ageing***

The rubber vulcanizates were thermally aged at  $90 \pm 1^\circ\text{C}$  for successive periods up to 7 days. The tensile strength, elongation at break and Young's modulus in the longitudinal and transversal directions were recorded. The data obtained are given in Table 3. From this table it is clear that the tensile strength of the control sample (without adhesion system [9]) decreases in both directions with increased ageing time, while it increases for samples containing adhesion system. This could indicate that the adhesion power increases with increased ageing time, which may be attributed to the formation of more crosslinked resin which improves power between SBR and PE short-fiber [6]. Also Young's modulus increased with increasing ageing time and carbon content in both directions, whereas elongation at break decreased with increasing ageing time and carbon content. This behavior could be attributed to further crosslinking in the rubber vulcanizates. It may be noticed that after thermal ageing, the elongation at break in the transversal direction is lower than in the longitudinal one for samples loaded with carbon black, which is the reverse of the relationship before ageing. This could confirm that some sort of degradation has happened during thermal ageing.

The retained values at each aging time were calculated using Eq. (1) and are given in Figure 2 for the longitudinal and transversal direction. This figure shows that the retained values of tensile strength of the control sample decrease with increased ageing time while it increases for the other samples with increasing both ageing time and carbon content. Retained values of elongation at break sharply decrease with ageing time and carbon content. While the values of Young's modulus increase with increased ageing time and decrease with increased carbon content. It is interesting to see that after 7 days thermal ageing, the retained value of Young's modulus for the sample loaded with 10 phr HAF increased up to 155% and 190% in the longitudinal and transversal directions respectively.

### ***Natural Ageing***

The previously investigated samples were exposed to normal weathering conditions for about two years. Then the mechanical properties were measured and listed in Table 4. From Tables 3 and 4 it is clear that the tensile strength, elongation at break and Young's modulus slightly increased during natural ageing. This indicates that these samples resist weathering conditions.



**FIGURE 2** The dependence of the retained values of the Tensile strength (T.S.) elongation at break and young's modulus on ageing time for SBR-PE fiber vulcanizates loaded with different contents of HAF carbon black. Control (●), 0 phr (+), 10 phr (□), 20 phr (x), 30 phr (◆), 40 phr (○), 50 phr (△) and 60 phr (\*).

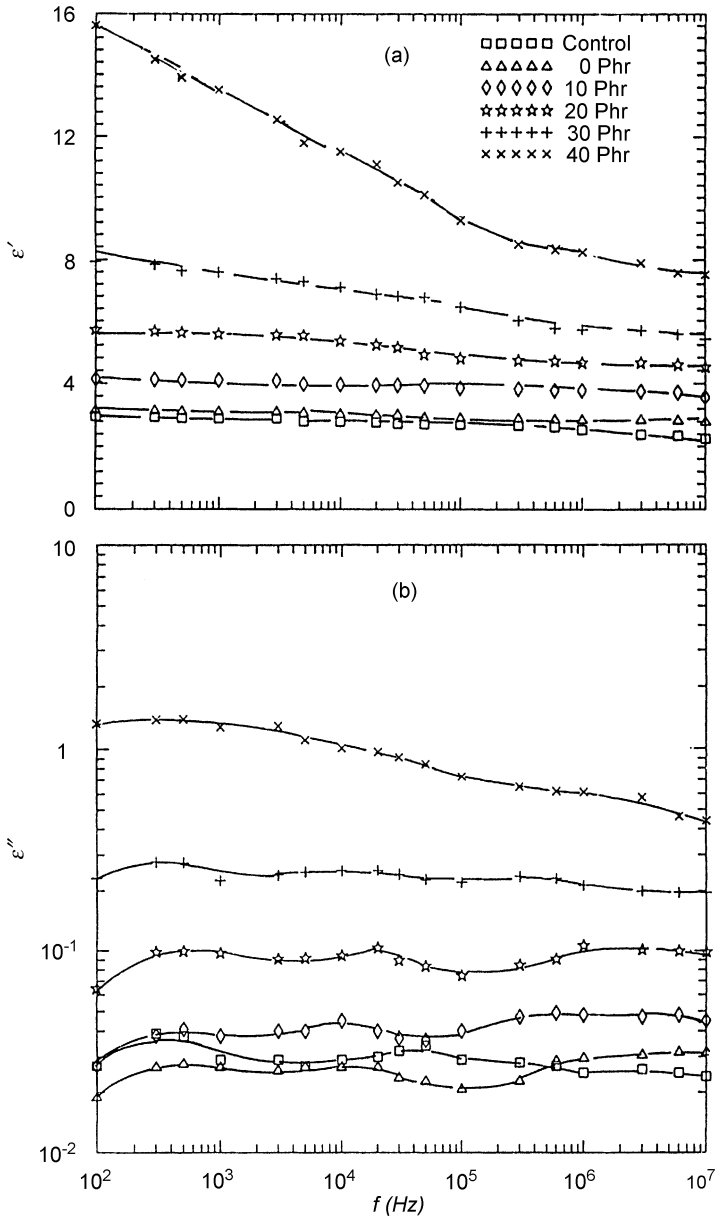
## Dielectric Properties

### *Effect of Frequency on the Dielectric Properties*

The permittivity  $\epsilon'$  and the dielectric loss  $\epsilon''$  for samples loaded with HAF black up to 40 phr were measured at a frequency range from 100 Hz to 10 MHz at 30°C. The data obtained are illustrated graphically in Figure 3. From Figure 3a it is clear that  $\epsilon'$  values increase with the increased carbon

**TABLE 4** Mechanical properties after two years natural ageing of SBR – PE fiber loaded with HAF carbon black in the Longitudinal (L) and Transversal (T) directions

<i>Property</i>	<i>Sample</i>	<i>Control</i>		<i>HAF black contents in (phr)</i>					
		<i>sample</i>	<i>0</i>	<i>10</i>	<i>20</i>	<i>30</i>	<i>40</i>	<i>50</i>	<i>60</i>
Tensile strength (N/mm <sup>2</sup> )	L	3.87	5.00	8.00	10.87	11.98	15.89	18.58	20.21
	T	3.87	4.46	6.27	8.42	9.20	13.00	13.95	15.89
Elongation %	L	543.00	186.21	163.00	153.90	144.00	122.00	119.00	100.81
at break	T	552.00	202.23	189.70	178.00	165.26	146.00	136.95	120.25
Young's modulus (N/mm <sup>2</sup> )	L	1.79	7.31	15.68	18.89	23.65	28.91	31.27	38.89
	T	1.28	6.28	12.51	14.21	18.81	22.11	26.81	30.16



**FIGURE 3** (a) The permittivity  $\epsilon'$  and (b) the dielectric loss  $\epsilon''$  versus frequency ( $f$ ) for SBR-PE vulcanizates loaded with different contents of HAF black (10, 20, 30 and 40) at 30°C.

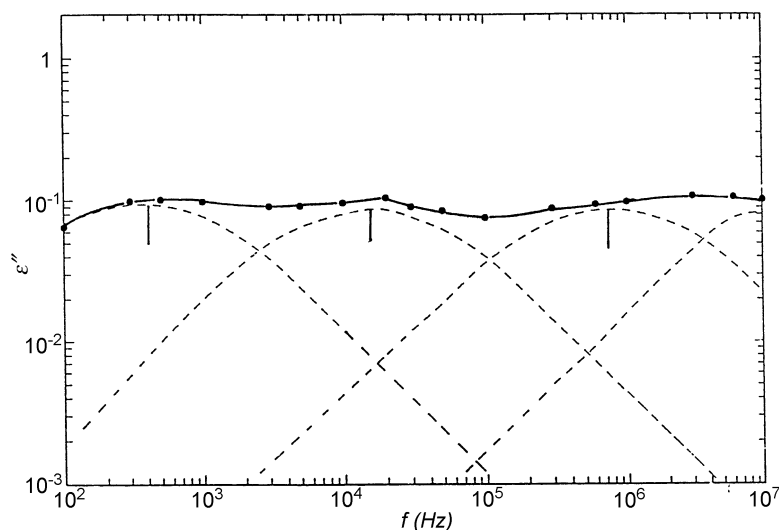
content and decrease with increased frequency which is more pronounced at high concentrations showing anomalous dispersion.

Figure 3b shows small variations of the dielectric loss  $\epsilon''$  with frequency at low concentrations up to 30 phr HAF black, while at 40 phr values of  $\epsilon''$  decrease with increased frequency. The absorption curves obtained are broad, indicating a number of relaxation processes. Similar behavior has been reported earlier [13–15].

The data were analyzed using a computer program based on superimposed Fröhlich terms [16]. Three absorption regions were obtained. Example of the analysis for sample having 20 phr HAF is shown in Figure 4.

The relaxation times  $\tau_i$ 's associated with the Fröhlich terms and the corresponding distribution parameters  $\rho_i$ 's for the samples under investigation are given in Table 5. It is seen from this table that values of  $\epsilon''_{max}$  increase with increased carbon content while the  $\rho_i$ 's are unchanged.

The relaxation times of the first absorption region ( $\tau_I \cong 4.5 \times 10^{-4}$  s) are about the same for all carbon contents. This absorption region could be due to Maxwell–Wagner losses [17–19] arising from interfacial polarization due to the presence of various ingredients added to rubber. These losses are due to an a.c. current which is in phase with the applied potential. This current results from the differences in conductivities and permittivities of the ingredients composing the vulcanized rubber samples. The relaxation times of the second absorption region ( $\tau_{II} \cong 10 \times 10^{-6}$  s) is also unchanged by the



**FIGURE 4** Absorption curves of SBR rubber–polyester fiber vulcanizate loaded with 20 phr HAF black. Fitting experimental  $\epsilon''$  values (●) Fröhlich terms (solid line).

**TABLE 5** Results of analysis of SBR-PE vulcanizates loaded with HAF black;  $\rho_i$  Fröhlich distribution parameter,  $\epsilon''_{max}$  value of maximum absorption  $\tau_i$  and relaxation time for the first (I), second (II) and third (III) absorption regions

<i>Carbon black content ( phr)</i>	$\rho_i$			$\epsilon''_{max}$			$\tau_i(s)$		
	I	II	III	I	II	III	I $\times 10^4$	II $\times 10^6$	III $\times 10^8$
<b>Before ageing</b>									
0	2.8	2.8	2.8	0.025	0.024	0.025	3.98	9.40	16.75
10	2.8	2.8	2.8	0.038	0.040	0.040	4.42	9.90	19.89
20	2.8	2.8	2.8	0.094	0.084	0.084	4.42	10.60	21.22
30	2.8	2.8	2.8	0.261	0.215	0.195	4.48	10.60	28.94
40	2.8	2.8	2.8	1.35	0.840	0.520	4.48	10.60	31.83
<b>After 7-days of thermal ageing</b>									
0	2.8	2.8	2.8	0.034	0.038	0.029	3.98	10.50	20.94
10	2.8	2.8	2.8	0.057	0.061	0.043	4.42	10.60	22.74
20	2.8	2.8	2.8	0.089	0.099	0.098	4.42	10.60	31.83
30	2.8	2.8	2.8	0.300	0.322	0.212	4.42	12.20	41.34
40	2.8	2.8	2.8	1.420	0.990	0.565	4.68	14.50	44.21

increase of carbon content. This may be attributed to either segmental mobility in the main chain or some sort of motion associated with the mobility of the main chain. The relaxation time of the third absorption region  $\tau_{III}$  is increased by the increase of carbon content from  $(16.7 \times 10^{-8} \text{ s})$  at zero concentration to  $(31.8 \times 10^{-8} \text{ s})$  at 40 phr HAF. This could be due to cooperative motions of the polar groups in the main chain arising from the addition of ingredients to the rubber matrix. It seems that polar groups in the main chain interact with the added carbon forming aggregates. The size of these aggregates did increase by further addition of carbon to the rubber matrix.

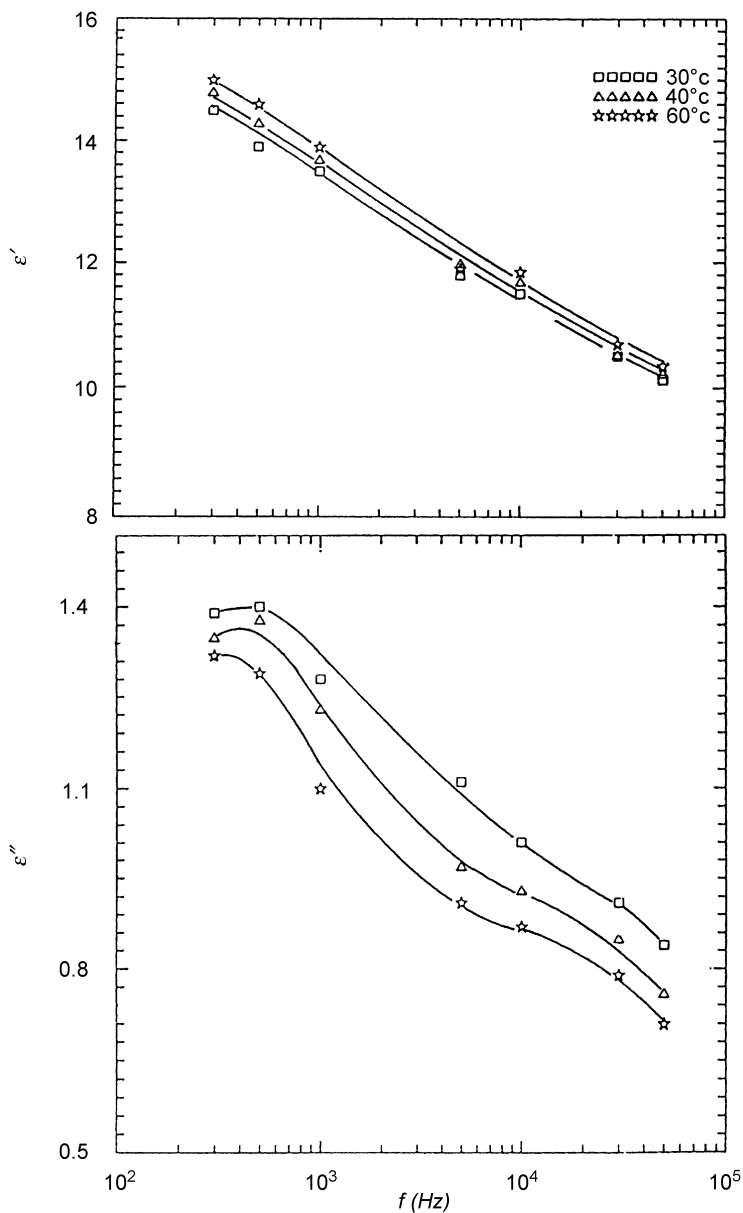
The values of the experimental data in the higher frequency range did not verify the point fitting analysis, so a fourth absorption region is expected in this frequency range, which is becoming more pronounced as the HAF content in the sample is increased. This region, although its maximum lies in a frequency range higher than the available one, can occur by segmental rotation or local twisting motion of the main chain.

### **Effect of Frequency at Different Temperatures on the Dielectric Properties**

The permittivity  $\epsilon'$  and the dielectric loss  $\epsilon''$  of the samples were measured in the frequency range between 100 Hz and 50 KHz at 30°C, 40°C and 60°C. No significant change in  $\epsilon'$  and  $\epsilon''$  with temperature was found for samples having low carbon concentrations. At higher concentration (40 phr)  $\epsilon'$  was found to increase slightly with temperature increase while  $\epsilon''$  decreased as is shown in Figure 5. This figure shows the dependence of  $\epsilon'$  and  $\epsilon''$  on frequency for the 40 phr HAF carbon black sample at temperatures 30, 40 and 60°C. It is clear that values of  $\epsilon'$  decrease with frequency and increase with temperature while  $\epsilon''$  decreases with both frequency and temperature. Two absorption regions are shown in the  $\epsilon''$ -log frequency curves, a low frequency one whose maximum is at frequencies lower than 1 KHz and another absorption region at higher frequencies whose peak is at about 15 KHz. As the temperature increases from 30 to 60°C the high frequency absorption region seems to become broader and shifts to lower frequencies. This may be attributed to an increase of the average relaxation times due to the orientation of either larger aggregates or crosslinked molecules formed by increasing the temperature.

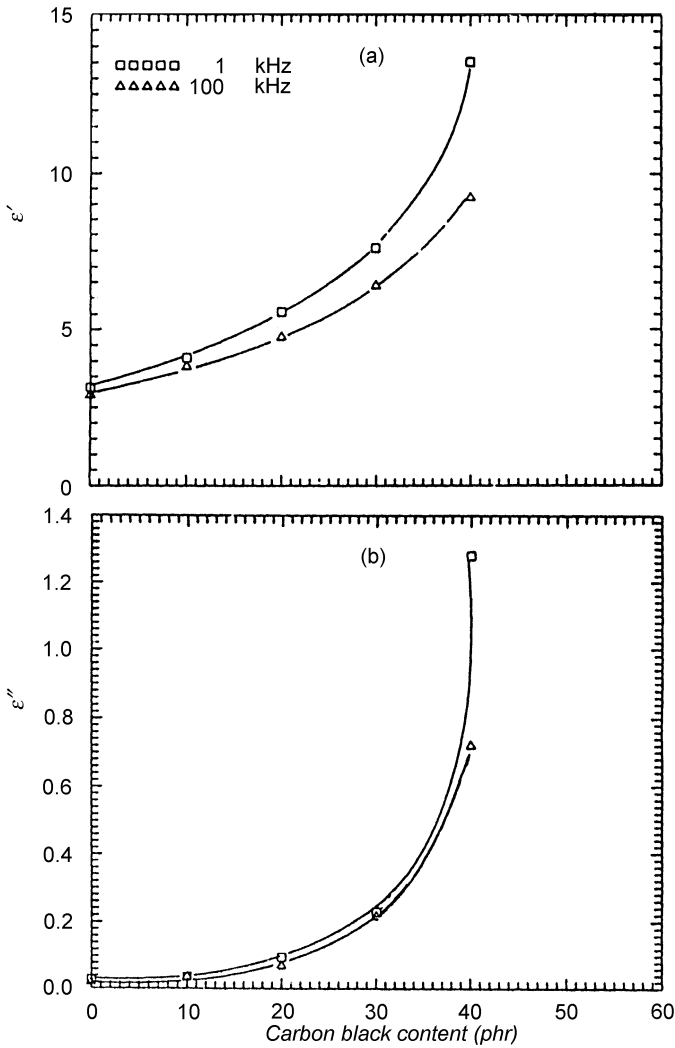
### **Effect of HAF Concentration on the Dielectric Properties**

Figure 6 shows the effect of HAF black content on the permittivity  $\epsilon'$  and the dielectric loss  $\epsilon''$  at 1 KHz and 100 KHz. It is clear that the values of  $\epsilon'$  and  $\epsilon''$  increase gradually with increased carbon content, except for 40 phr



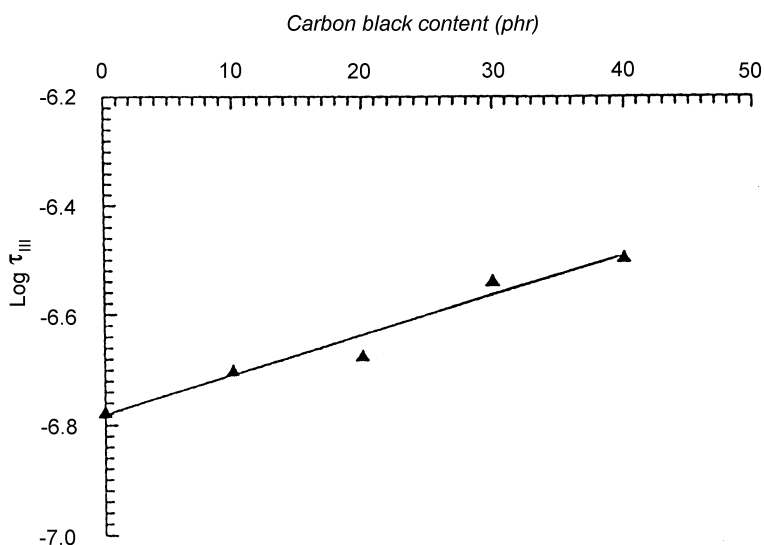
**FIGURE 5** The permittivity  $\epsilon'$  and the dielectric loss  $\epsilon''$  versus frequency ( $f$ ) for SBR-PE fiber vulcanizate loaded with 40 phr HAF at different temperatures.





**FIGURE 6** Dependence of the (a) dielectric permittivity  $\epsilon'$  and (b) dielectric loss  $\epsilon''$  on the carbon black (HAF) content.

HAF where there is an abrupt increase. This abrupt increase could be due to the onset of dc conductivity when infinite conducting chains are formed inside the insulating matrix. Above 40 phr, aggregates begin to come in contact with each other. This causes a sharp increase in the physical properties [15, 20–23]. When converted to volume fractions, the onset of



**FIGURE 7** Variation of  $\log \tau_{III}$  with carbon black content for SBR-PE fiber vulcanizates samples loaded with HAF carbon black.

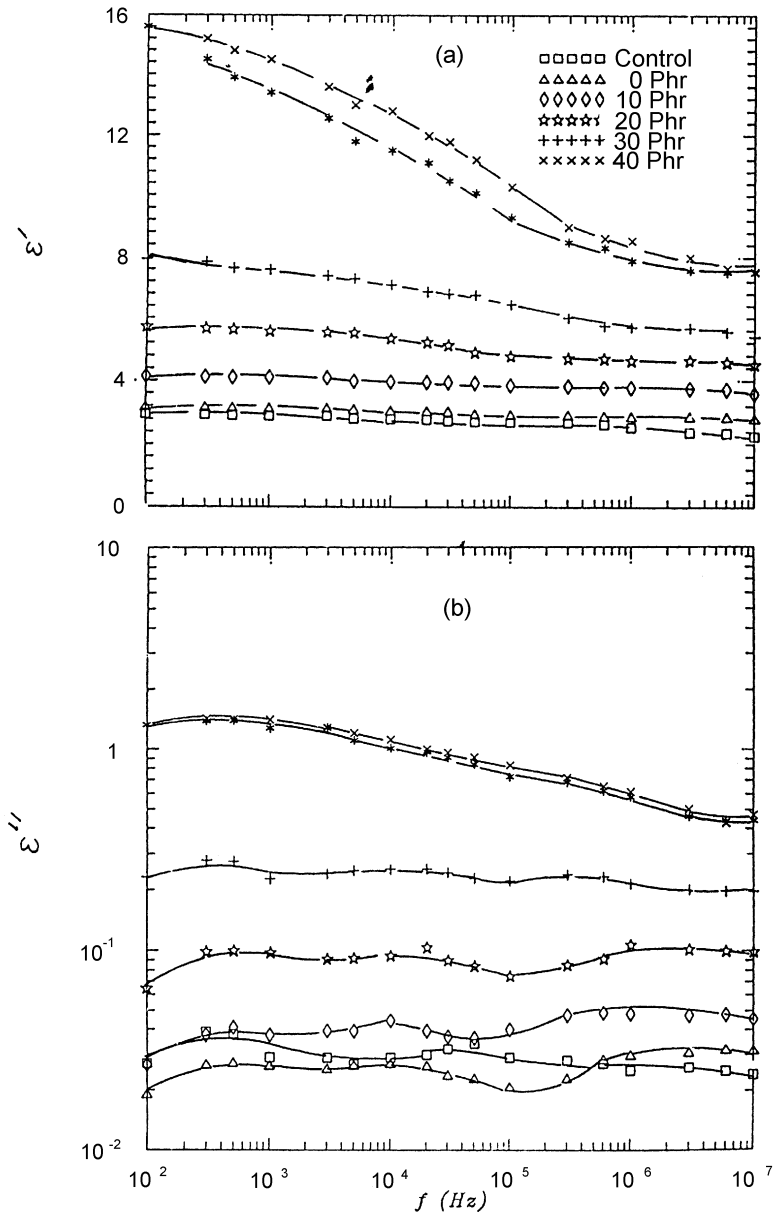
conductivity at 40 phr carbon is in excellent agreement with the expectations from Percolation Theory [24].

Plotting  $\ln \tau_{III}$  as a function of carbon content Figure 7, a linear relationship with a slope  $\cong 0.7$  is obtained. This result agrees with that found before by Schallamach and others [25,26] Schallamach found that the dielectric relaxation time is approximately an exponential function of the percentage of combined sulfur. Thus as in case of sulfur, the relation between the dielectric relaxation time and carbon content is exponential. This figure can be used to find out the relaxation time for any other concentration.

## Effect of Ageing on the Dielectric Properties

### Thermal Ageing

The investigated samples were exposed to thermal ageing at  $90 \pm 1^\circ\text{C}$  for 7 days. The dielectric properties were remeasured at  $30^\circ\text{C}$ , comparing these data with those obtained before ageing. It was found that the values of  $\epsilon'$  for the control sample slightly decreased, while those of  $\epsilon''$  increased. For the other samples  $\epsilon'$  and  $\epsilon''$  both increased. The obtained dielectric data after thermal ageing were analyzed as before with the results given in Table 3. From this table, it is clear that the relaxation times of the first absorption region are about the same as before ageing, while those of the second absorption region increased at the high concentrations. On the other hand,



**FIGURE 8** (a) The permittivity  $\epsilon'$  and (b) the dielectric loss  $\epsilon''$  versus frequency ( $f$ ) for naturally aged SBR-PE vulcanizates loaded with different contents of HAF black (10, 20, 30 and 40) at 30°C. \* 40 phr after heating.

the relaxation times of the third absorption region increased for all samples after thermal ageing. This may be attributed to the formation of larger aggregates and/or formation of more crosslinked molecules which is expected to take place during thermal ageing. This is supported by the increase in tensile strength and Young's modulus after thermal ageing [27].

### **Natural Ageing**

$\epsilon'$  and  $\epsilon''$  were measured after two years of naturally aged samples, the data obtained are illustrated graphically in Figure 8. Comparing these results with those obtained before ageing (Fig. 3), no significant change in  $\epsilon'$  and  $\epsilon''$  is noticed for low concentrations up to 30 phr HAF carbon black. For concentration 40 phr HAF black  $\epsilon'$  and  $\epsilon''$  slightly increased. The increase in  $\epsilon'$  and  $\epsilon''$  may be due to the absorption of moisture by the sample from the environment. Similar behavior was found before [28, 29]. To check on this, the sample was heated at 70°C under vacuum (to avoid deterioration of the rubber sample) for about 24 hours, then the permittivity  $\epsilon'$  and the dielectric loss  $\epsilon''$  were remeasured and it was found that they indeed decreased. So the increase in  $\epsilon'$  and  $\epsilon''$  is mostly due to moisture absorbed by the sample, and the values before ageing are regained by the sample.

### **CONCLUSIONS**

- (1) Tensile strength and Young's modulus of SBR – PE composites loaded with HAF carbon black increased by increasing carbon content while elongation at break decreased. Also these properties showed the same trend with ageing time.
- (2) Values of tensile strength and Young's modulus in the longitudinal direction were higher than the transversal one while an opposite trend was found for elongation at break. This indicates that the fibers were inside the rubber matrix fairly oriented in the longitudinal direction.
- (3) The retained values of tensile strength increased with increased ageing time and carbon content whereas those of Young's modulus increased with increased ageing time and decreased with the increase of carbon content. This could be attributed to the formation of more crosslinked resin which improve adhesion power between SBR and PE fiber.
- (4) At 40 phr HAF there was an abrupt increase in  $\epsilon'$  and  $\epsilon''$  suggesting the formation of conducting chains inside the insulating matrix, as expected from Percolation Theory.
- (5) Three absorption regions were obtained from the analysis of the dielectric absorption spectra. The first one was, due to Maxwell–Wagner effect, the second could be due to segmental mobility in the main chain and the third one may be due to cooperative motion of the polar groups in the main chain.

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