Newsprint Paper Waste as a Fiber Reinforcement in Rubber Composites

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ABSTRACT: The investigation of the economical use of lignocellulose waste, which is one of the environmental problems facing third-world nations, is ongoing. In this study, we intended to increase the use of newsprint fibers in the rubber industry. For this reason, we treated newsprint fibers with sodium silicate and magnesium chloride, and we examined the water retention values and thermal degradation analyses of the treated fiber waste. The activation energy of degradation was evaluated with the Coats–Redfern method of analysis. The effects of the addition of different amounts [0–60 parts per hundred parts of rubber by weight (phr)] of untreated and treated newsprint fibers on both the electrical and mechanical properties of two different types of rubber (natural rubber and acrylonitrile rubber) were studied. This study led us to the conclusion that the addition of treated newsprint fiber waste at a concentration of 40 phr could lead to an end product characterized by good electrical and mechanical properties. © 2003 Wiley Periodicals, Inc. J Appl Polym Sci 91: 469–478, 2004

Key words: fibers; reinforcement; rubber; composites

INTRODUCTION

Newer materials and composites with both economic and environmental benefits are being considered for applications in the automotive, building, furniture, and packaging industries. Mineral fillers and fibers are used frequently in the plastics industry to achieve desired properties or to reduce the cost of the end products. Recent research into the use of annual-growth lignocellulose fibers suggests that these fibers have the potential for use as reinforcing fibers in thermoplastics.[1–4] The primary advantages of using annual-growth lignocellulose fibers as additives in plastics are the low density, low cost, nonabrasive nature, high filling levels possible, low energy consumption, and wide variety of fibers available throughout the world. The two main disadvantages of using lignocellulose fibers in thermoplastics are the high moisture absorption of the fibers and composites and the low processing temperatures permissible.[5,6] When paper waste is used as secondary fibers for paper making, it requires extensive stages for deinking, cleaning, and refinement. When paper waste, especially newsprint paper, is used for the manufacture of composites, it does not require extensive preparation. This greatly reduces the potential cost of manufacturing.[7,8] Our earlier work on newspaper waste recommended its use as inexpensive agro fibers for the production of lightweight buildings in interior application panels.[9,10]

Studies on the effects of black fillers on the dielectric and physicomechanical properties of natural rubber (NR),[11] perbunan rubber,[12] SBR,[13,14] ethylene–propylene–diene monomer,[15] butyl rubber,[16] and neoprene rubber[17] have been carried out.

The effects of white fillers on the electrical and physicomechanical properties of NR,[18] butyl rubber,[19] perbunan rubber,[20] and SBR[21] have been studied.

Shin Zo and Yuko[22] determined that silica was a good reinforcing agent because it improved both the tensile and dynamic mechanical parameters. Lary and Huber[23] showed that the presence of mineral fillers and silica in rubbers improved compound formulations for tire products, adhesives, and coatings.

Resorcinol adhesives have been found to be outstanding not only for their excellent durability but also for their unique property of rapid curing without heat and their high-strength bonds. Therefore, they can be used in the bonding of many materials, such as wood, paper, textiles, plastics, rubber, metals, glass, ceramics, and concrete. The adhesion between many types of materials and most elastomers has been overcome by the discovery of a three-component system (HRH)[24–26] of hydrated silica, resorcinol, and hexamethylene tetramine (HMT). The theory of adhesion can be explained by the *in situ* formation of resorcinol formaldehyde in rubber vulcanizates.[27]

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NR is considered a good insulating material because it has good mechanical properties due to its crystallization by orientation.[23] The aim of this investigation was to study the effects of newsprint fiber waste, considered to be a source of environmental pollution, before and after treatment with sodium silicate and magnesium chloride as fillers in increasing quantities on both the electrical and mechanical properties of NR.

Butadiene acrylate copolymer (NBR) is characterized by good mechanical properties and good oil resistance,[28] but it has moderate insulating properties despite the high value of its permittivity ($\epsilon' = 17$–14).[29] The considerably high dielectric loss ($\epsilon'' = 1.33$–2.00) to some extent deprives NBR of its insulating properties. For this reason, we aimed to load it with newsprint fiber waste to reduce the higher values of ϵ'' and keep the changes in both ϵ' and the mechanical properties to a minimum.

Silica is usually used as a reinforcement material and always give better properties.[19,20] Therefore, we aimed to compare the properties of rubber products obtained from newsprint fiber waste with those obtained from silica in an attempt to replace silica with newsprint fiber waste in fiber-reinforced rubber composites.

EXPERIMENTAL

Materials

NR (SMR-20) was acquired in the form of ribbed, smoked sheets with a specific gravity of 0.913, a Mooney viscosity at 100°C of 60–90 r, and a glass-transition temperature of −75°C.

NBR was obtained with an acrylonitrile content of 32% and a specific gravity of 1.17.

Zinc oxide and stearic acid were used as activators with specific gravities at 15°C of 5.55–5.61 and 0.9–0.97, respectively. The plasticizer was naphthenic processing oil with a specific gravity of 0.94–0.96, and the accelerator was *N*-cyclohexyl-2-benzothiazole sulfenamide (CBS) with a specific gravity of 1.24–1.32 and a melting point of 95–100°C. The antioxidant was phenyl-β-naphthyl amine (PBN) with a specific gravity of 1.18–1.2 and a melting point of 105–106°C. The curing system was sulfur powder with a specific gravity of 2.04–2.06.

Resorcinol, HMT, and hisil were pure grades obtained from El-Nasr Pharmaceutical Chemicals Co.

Light reinforcing silica (82% precipitated silicon dioxide) with a specific gravity of 1.95 was used as a filler.

Newsprint paper was used as a reinforcement for rubber composites. The chemical and physical analyses of the fiber waste were carried out according to standard methods[30–33] and were recorded in Table I.

TABLE I
Chemical and Physical Properties of the Newsprint Paper Fiber Waste

Property	Untreated	Treated
Klason lignin (%)	21.7	—
α-Cellulose (%)	64.54	—
Pentosans (%)	12.39	—
Ash (%)	7.32	—
Water retention value (%)	159.5	84.3

The treatment of the newsprint fibers was carried out as follows: the ground fibers were mixed in a kneader with 20% sodium silicate (with respect to the fibers; the molar ratio of SiO_2 to Na_2O was 3.5:1) for 2 min, an equivalent amount of magnesium chloride was added with constant mixing for 3 min.

Mixing

The mixing was carried out on a laboratory two-roll mill (470-mm diameter and 300-mm working distance). The speed of the slow roll was 24 rpm with a gear ratio of 1:1.4. The compounded blends were left overnight before vulcanization.

Vulcanization

The vulcanization was carried out in a heated platinum press under a pressure of about 40 kg/cm^2 and at a temperature of 152 ± 1°C.

Techniques

Thermal analysis

The fire retardancy of the treated recycled newsprint fibers was examined with thermogravimetric techniques. Thermogravimetry (TG) and derivative thermogravimetry (DTG) of the untreated and treated newsprint fibers were performed with a PerkinElmer TGA7 thermogravimetric analyzer. The analysis was performed at a heating rate of 10°C/min and a flow rate of 50 cc/min, under nonisothermal conditions, in the presence of nitrogen.

The determination of the activation energy (E_a) from data of the weight loss versus the temperature was carried out with the Coats–Redfern equation.[34] The general correlation equation is

$$\log_{10}\left[\frac{1-(1-\alpha)^{1-n}}{T^2(1-n)}\right] = \log_{10}\left[\frac{AR}{aE_a}\left(1-\frac{2RT}{E_a}\right)\right] - \frac{E_a}{2.3RT} \quad (1)$$

where α is the fractional conversion, n is the order of the reaction, a is the heating rate (K min^{-1}), R is the gas constant (kJ mol^{-1}), T is the temperature (K), and A is the frequency factor (S^{-1}).

Plotting $\log_{10}[1 - (1 - \alpha)^{1-n}/T^2(1 - n)]$ against $1/T$ with different values of n should give a straight line with the most appropriate value.[35] Thus, the method of least squares was applied to the equation: n ranged from 0 to 3.0 in increments of 0.5, and for each value of n, the correlation coefficient ($-r$), standard error estimation (SE), and E_a were calculated. n, which corresponded to maximum $-r$ and minimum SE values, was the order of the degradation process. E_a was calculated from the slope.

Rheometric characteristics

The minimum torque (M_L), the maximum torque (M_H), the optimum cure time at 90% of the maximum torque (T_{C90}), the scorch time (T_{S2}; i.e., the time taken for M_L to increase by two units), and the cure rate index (CRI) were determined according to ASTM D 1646 1994 with a Monsanto 1009 oscillating rheometer.

Mechanical properties

The stress and strain at yield and rupture were determined at room temperature on a Zwick 1425 tensile testing machine according to ASTM D 412-98a 1998.

Dielectric measurements

The dielectric measurements were carried out from 100 Hz to 100 kHz with an AG-411 B LCR meter (Ando Electric, Ltd., Japan). The capacitance and the loss tangent were obtained directly from the bridge from which ϵ' and ϵ'' were calculated. A guard ring capacitor (NFM/5T, Wiss Tech. Werkstatten GmbH, Germany) was used as a measuring cell. The cell was calibrated with standard materials,[36] and the experimental errors in ϵ' and ϵ'' were found to be ±3 and ±5%, respectively.

RESULTS AND DISCUSSION

Properties of the waste paper

From Table I, it is clear that the treatment of fiber waste with sodium silicate in combination with magnesium chloride improves the water resistance by approximately 47% with respect to untreated newsprint fibers.

Thermogravimetric analysis

The nonisothermal TG and DTG curves of untreated and treated newsprint fibers are illustrated in Figure 1. Figure 2 shows a plot of $-r$ and SE as functions of n for the decomposition stages of the examined newsprint

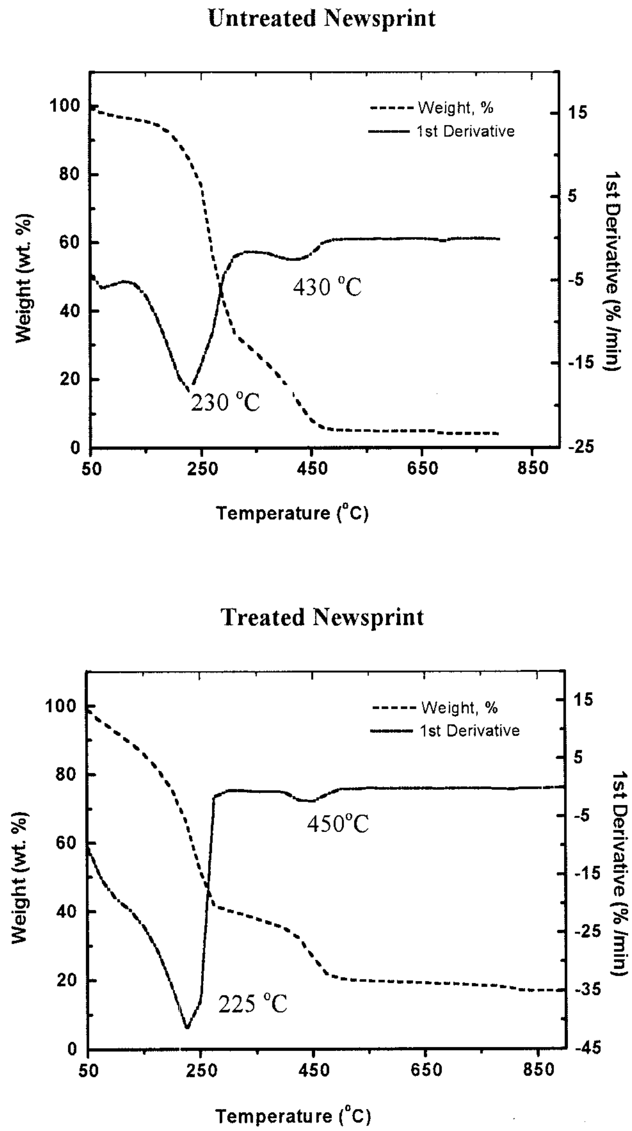

Figure 1 TG and DTG curves of untreated and treated newsprint waste paper.

samples to calculate the n and consequently E_a of degradation. Table II summarizes the temperature range, DTG peak temperature, $-r$, SE, n, E_a, and weight remaining of two main degradation stages of untreated and treated fiber waste.

From Figure 1, it is clear that after the initial loss of moisture at 50–110°C, the untreated newsprint fibers begin to degrade, and they lose weight at about 130°C. The DTG curve shows the rate of weight loss peaking at 230°C. The second degradation stage begins at 330°C, and the temperature exceeds 490°C before the weight loss of the untreated sample nears completion. The peak maxima of the DTG curve of the second degradation stage appear at 450°C.

Newsprint fibers treated with sodium silicate and magnesium chloride show a difference in the thermal degradation in comparison with untreated newsprint.

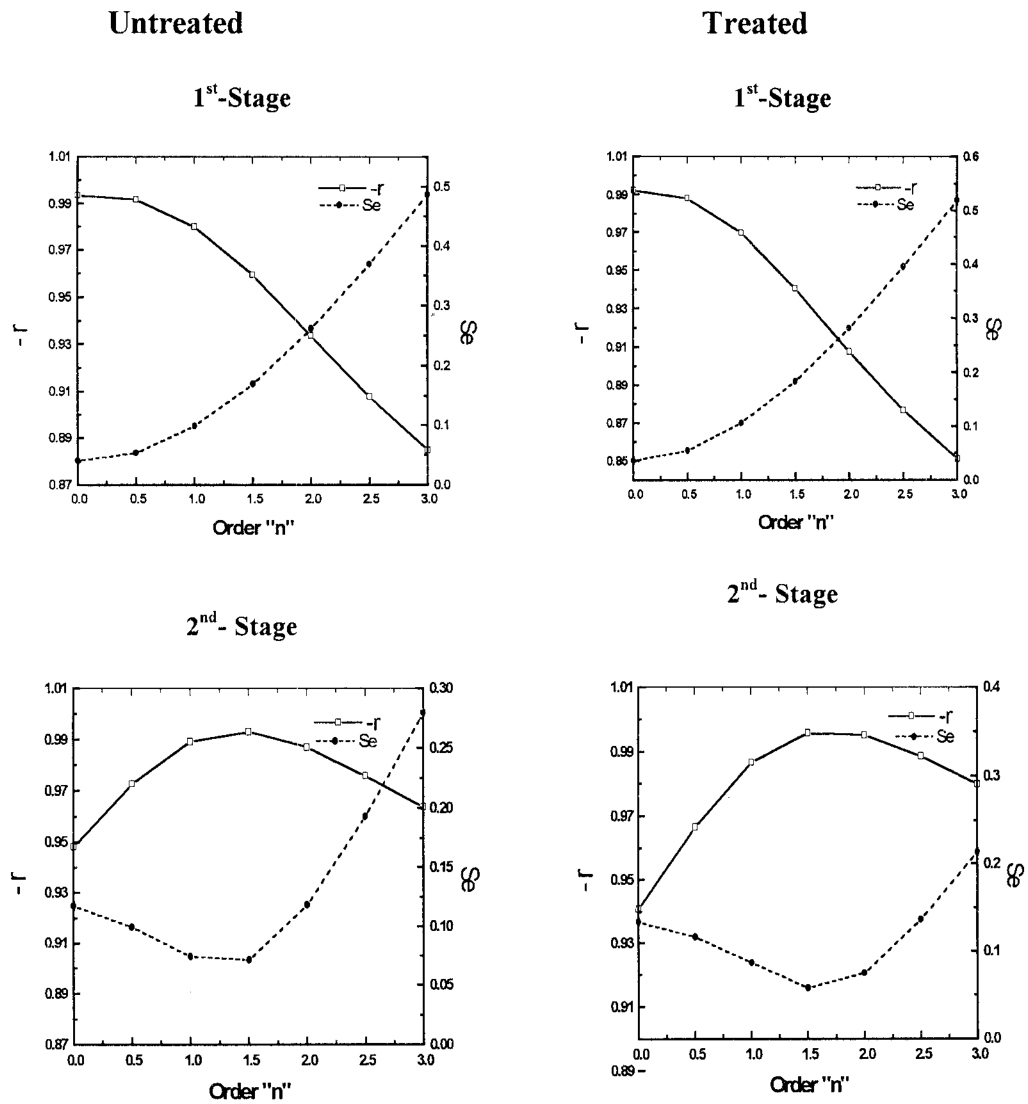

Figure 2 Statistical determination of n for the main degradation stages of untreated and treated newsprint fibers.

The weight loss of the first main degradation stage begins at about 50°C, and the thermal degradation is rapid and peaks at 225°C, which is lower than the peak temperature for untreated fiber waste. In contrast, for untreated newsprint, 66.8% of the weight loss occurs at the end of the first main degradation stage (at 310°C). The treatment of newsprint fibers reduces the weight loss of the first degradation stage to approximately 58% and diminishes E_a of the degradation stage from 47.39 to 16.34 kJ/mol. Treated newsprint

TABLE II
Measurements of TG and DTG Curves of the Main Degradation Stage of Untreated and Treated Fibers

Measurement	Untreated newsprint fibers		Treated newsprint fibers	
	First stage	Second stage	First stage	Second stage
Temperature range (°C)	130–310	330–490	50–275	400–500
DTG peak temperature	230	430	225	450
$-r$	0.99367	0.99309	0.9925	0.9958
SE	0.04032	0.07176	0.03600	0.0580
n	0.0	1.5	0.0	1.5
E_a (kJ/mol)	47.3890	125.105	16.340	262.369
Weight remaining (wt %)	33.252	5.304	41.757	20.413

TABLE III
Formulation of NR Mixes

Ingredient	phr
NR	100
Stearic acid	2
Zinc oxide	4
PBN	1
CBS	1
Sulfur	2
Oil	3
Filler (silica or waste paper either treated or untreated)	0–60

fibers have 20.4% residual char, whereas untreated fibers have 5.3% residual char, at 500°C.

The increase in the amount of the residual char and other changes in the thermal degradation caused by the treatment of the newsprint fibers (a higher rate of dehydration than of degradation in the first stage) are indicative of the effective flame resistance of treated newsprint fibers.

In light of this investigation, it is worthwhile to describe the potential of using untreated newsprint paper and newsprint paper treated with sodium silicate and magnesium chloride as fiber reinforcements in rubber composites.

Properties of the rubber composites with newsprint fibers as reinforcements

Two different types of rubbers were chosen: a nonpolar one, NR, which was characterized by its crystallization by orientation,[23] and a polar one, synthetic NBR, with the aim of investigating the electrical and mechanical properties of the prepared rubber composites.

NR

The basic formulation used for preparing the NR samples is given in Table III.

Table IV summarizes the rheometric characteristics and mechanical properties of the prepared samples.

The M_L values, which are thought to be a measure of the extent of mastication, slightly decrease with increasing amounts of silica and treated newsprint fiber waste, whereas no significant change in M_L has been noticed with untreated waste. M_H is an index of the crosslinking density, and its values indicate that the crosslinking density increases with increasing filler content; its highest values are recorded for untreated waste newspaper samples, especially for 20 parts per hundred parts of rubber by weight (phr). T_{S2} is a measure of the premature vulcanization of the material. The T_{S2} values in Table IV indicate that the samples loaded with treated newsprint fiber waste exhibit better scorch safety. T_{C90} is the optimum cure time for vulcanization and is calculated as follows:

$$T_{C90} = (M_H - M_L) \times 0.9 + M_L \quad (2)$$

It is clear that T_{C90} values generally decrease with the addition of the filler. CRI is a direct measure of the fast curing nature of the rubber compounds and is calculated with the following relation:

$$\mathrm{CRI} = 100/T_{C90} - T_{S2} \quad (3)$$

It is clear from Table IV that all the mechanical properties (stress at yield, stress at rupture, strain at yield, and strain at rupture) increase with the addition of 20 phr filler and then decrease with increasing filler content. This means that the optimum concentration for such loaded fillers is 20 phr.

ϵ' and ϵ'' were determined for NR loaded with different ratios of newsprint fiber waste (untreated and treated) and with silica in increasing quantities (0–60 phr). The measurements were carried out at room temperature (≈25°C), and the data are given in Figure 3. ϵ' increases with increasing filler content and decreases with increasing applied frequency, showing an anomalous dispersion. This increase is much more pronounced for treated newsprint fibers, as shown in a plot of ϵ' versus the filler content at a fixed frequency of 100 Hz [Fig. 4(a)]. The increase in ϵ' with increasing filler content may be due to the presence of polar groups in the filler. This increase occurs in the following order: treated newsprint fiber waste > silica > untreated newsprint fiber waste.

The variation of ϵ'' with the applied frequency (Fig. 3) indicates a pronounced increase, especially at a higher content of the filler.

From Figure 3, it is clear that the curves relating ϵ'' and log f are broader than the Debye curve,[37] indicating that more than one relaxation process is present. These processes could be attributed to the relaxation mechanisms of the main chain and its related motions.[15–17] One of the expected mechanisms is related to the Maxwell–Wagner effect, which usually occurs at a lower frequency range for heterogeneous systems. The origin of this process is an alternating current that is in phase with the applied potential because of the differences in the ϵ' and resistivity values of the constituents. Moreover, the shape of the spectra is unchanged by the addition of fillers because no interaction is expected to take place between the fillers and rubber. ϵ'' increases in the following order: treated waste newspaper > untreated waste newspaper > silica. This is clear from Figure 4(a).

It is worthwhile to study the effects of adding the different constituents comprising the HRH system, which gives good results for the adhesion of short fibers and most elastomers[38] with respect to the me-

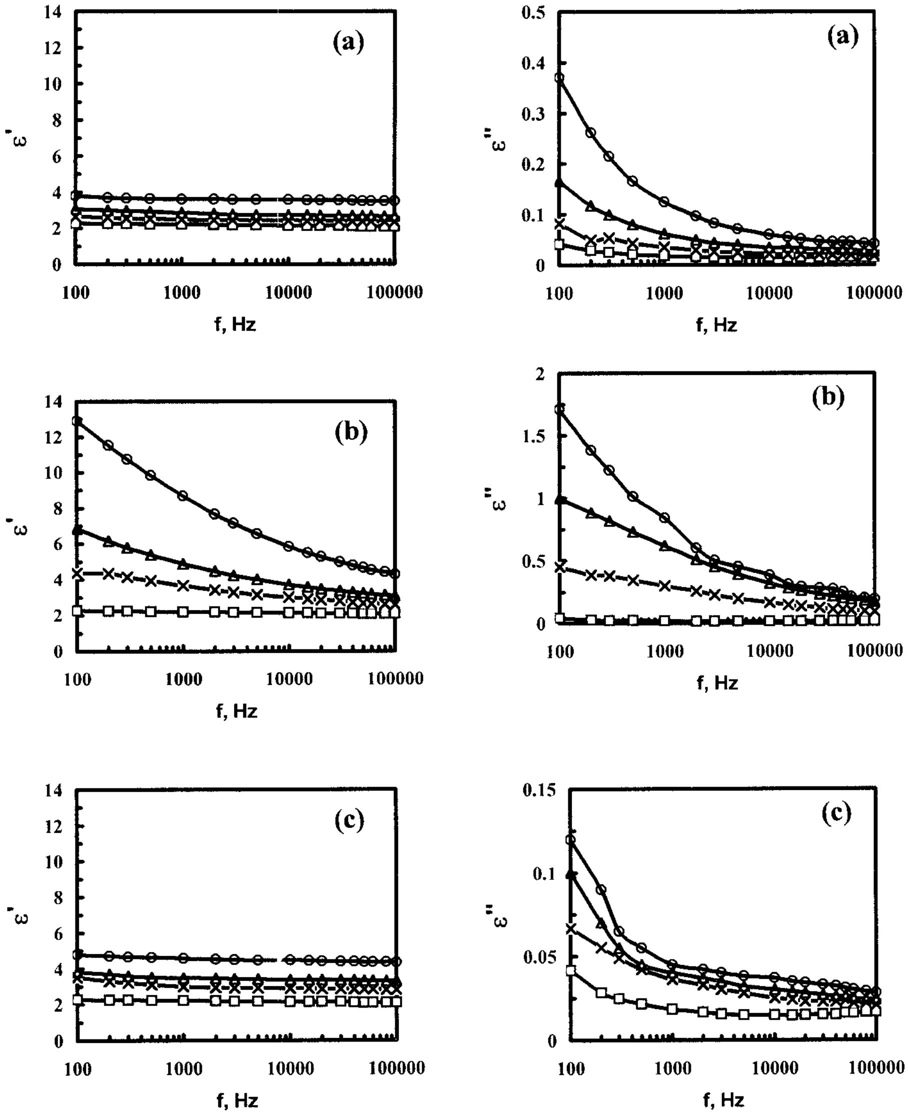

Figure 3 ε′ and ε″ of NR loaded with (a) untreated newsprint waste, (b) treated newsprint waste, and (c) silica at room temperature (25°C): (□) 0, (×) 20, (△) 40, and (○) 60 phr.

chanical and dielectric properties of the investigated samples.

The basic formulation used for preparing NR samples is similar to that given in Table III (plus 5 phr resorcinol and hydrated silica and 3.2 phr HMT).

The rheometric characteristics and mechanical properties of the samples are summarized in Table V. Comparing Tables IV and V, we find that the rheometric characteristics and mechanical properties of the samples loaded with silica are slightly enhanced, so there is no need for the adhesion system for such samples. However, a pronounced increase in M_H followed by a decrease in T_{C90} can be seen for those loaded with newsprint fiber waste either treated or untreated. The stress at yield and at rupture and the strain at yield and at rupture are highest for the samples loaded with 20 and 40 phr treated newsprint fiber waste; this means that the adhesion system increases the maximum loading concentration and gives optimum mechanical properties up to 40 phr.

ε′ and ε″ were measured in the way discussed previously, and the obtained data are illustrated graphically in Figure 5. Both ε′ and ε″ increase with increasing filler content and decrease with increasing frequency. ε′ follows this order: treated newsprint waste > untreated newsprint waste > silica. ε″ follows this order: treated newsprint waste < silica < untreated newsprint waste. These results recommend treated newsprint waste/NR composites for use in insulation.

Figure 4(b) presents the variations of ε′ and ε″ at a fixed frequency of 100 Hz. Comparing this figure with that for samples without HRH [Fig. 4(a)], we find that for untreated samples, both ε′ and ε″ increase for the loaded samples in comparison with those lacking the HRH system. For treated samples, ε′ slightly increases up to 40 phr, whereas a significant decrease in ε′ can

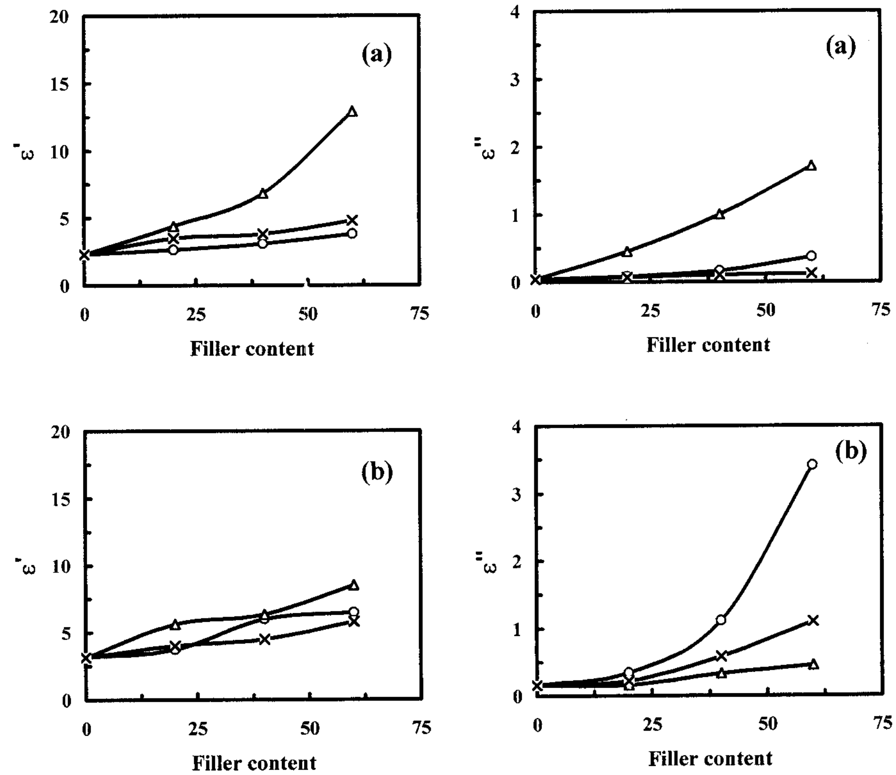

Figure 4 ϵ' and ϵ'' at 100 Hz of (a) NR without the adhesion system (HRH) and (b) NR with the adhesion system loaded with different contents of (○) untreated newsprint waste, (△) treated newsprint waste, and (×) silica.

be observed at 60 phr with the addition of the HRH system. However, an increase in ϵ'' can be noticed with the addition of the HRH system only for the unfilled samples, whereas a pronounced decrease in ϵ'' can be noticed after the addition of treated newsprint waste (20–60 phr). For silica samples, it is clear that ϵ' slightly increases, whereas ϵ'' greatly increases, with the addition of HRH system.

From this investigation, it can be concluded that treated newsprint fiber waste can be used as a filler instead of silica, but the concentration should not exceed 40 phr.

NBR rubber

NBR is characterized by good mechanical properties and moderate insulating properties because it possesses high values of ϵ' (17–14)[29] and very high ϵ'' values (1.33–2.00) that, to some extent, lessen its insulating properties. For this reason, we loaded it with untreated and treated newsprint fiber waste and silica in the same manner mentioned previously for NR with the aim of reducing ϵ'' and keeping the changes in both ϵ' and the mechanical properties to a minimum.

The basic formulation used for preparing the NBR samples is given in Table VI.

The rheometric characteristics and mechanical properties of the samples are summarized in Table V. M_H increases with increasing filler content up to 40 phr, and then a slight decrease can be observed; this increase is followed by a decrease in T_{C90} values, which suggests optimum condition at a 40 phr loading. M_H is highest and T_{C90} is lowest for a sample loaded with 40 phr treated waste paper, which is highly recommended for mechanical purposes. These results find further justification in the stress at yield and at rupture and in the strain at yield and at rupture.

ϵ' and ϵ'' were determined for NBR loaded with different ratios of newsprint fiber waste (untreated and treated) and silica in increasing quantities (0–60 phr). The measurements were carried out at room temperature (≈25°C). The obtained data are illustrated graphically in Figure 6. ϵ' decreases with an increasing applied frequency, showing an anomalous dispersion. In this range, ϵ' has contributions from orientation and atomic and electronic polarizations.[28] Figure 7 presents the variations of ϵ' with the filler content. ϵ' increases with increasing filler content. As the concentration of the treated newsprint waste increases up to 60 phr, an unpronounced change in ϵ' can be noticed. This can be attributed to the increase in the density of the system, which leads to a reduction in the orientation of the dipole in comparison with those with a lower content of the filler (20–40 phr).

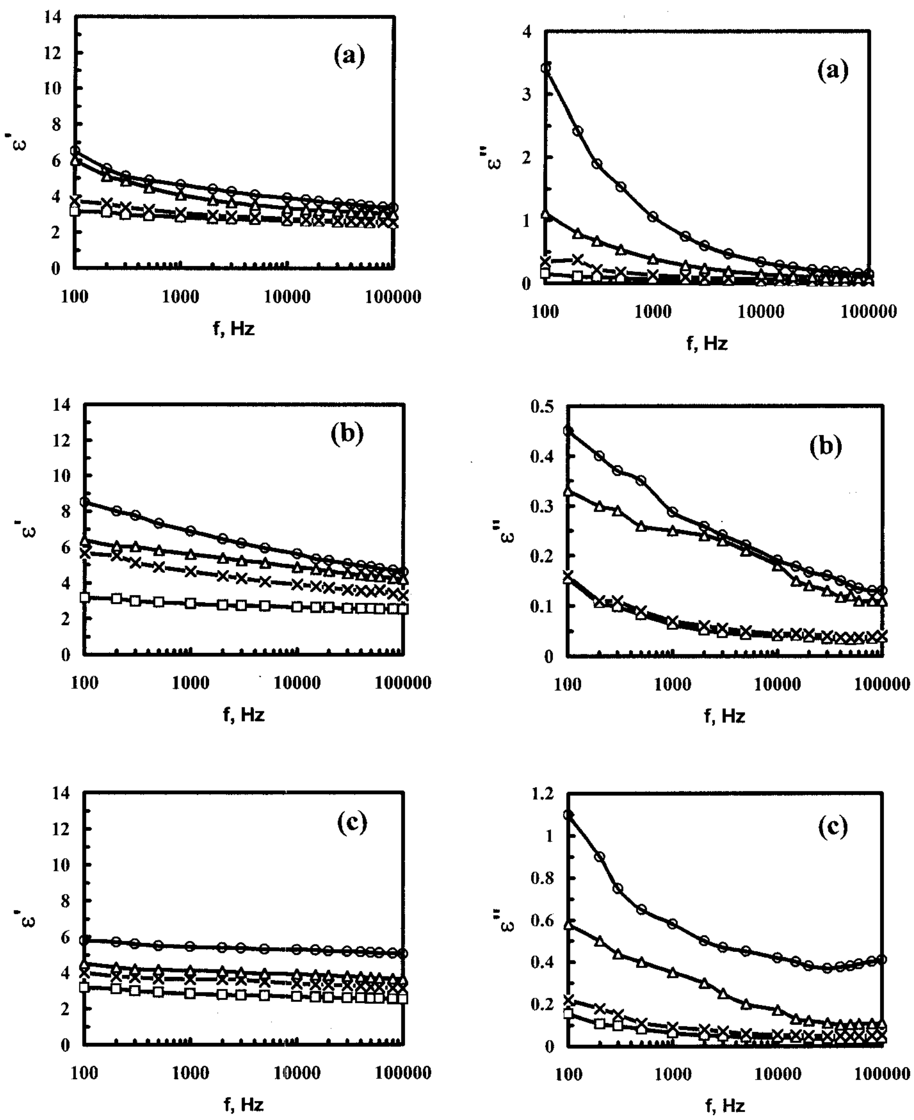

Figure 5 ϵ' and ϵ'' of NR with the adhesion system (HRH) loaded with (a) untreated newsprint waste, (b) treated newsprint waste, and (c) silica at room temperature (25°C): (□) 0, (×) 20, (△) 40, and (○) 60 phr.

The curves relating ϵ'' and $\log f$ (Fig. 6) are broader than the Debye curve,[25] indicating that more than one relaxation process is present. These processes can be attributed to mechanisms related to the main chain and its related motions.[15–17] One of the expected mechanisms is related to the Maxwell–Wagner effect, which usually occurs at the lower frequency range for heterogeneous systems.

TABLE VI
Formulation of NBR Mixes

Ingredient	Phr
NR	100
Stearic acid	2
Zinc oxide	4
PBN	1
CBS	1
Sulfur	2
Oil	3
Resorcinol	5
Hydrated silica	5
HMT	3.2
Filler (silica or waste paper either treated or untreated)	0–60

Figure 7, which summarizes the variation of ϵ'' with the filler content, shows a pronounced decrease in ϵ'' with increasing filler content up to 40 phr for both treated and untreated newsprint fibers and up to 60 phr for silica. This decrease may be due to the hindrance in the orientation of C≡N, which is expected to take place in the presence of the filler. At a higher loading (60 phr), a significant increase in ϵ'' can be noticed for the treated newsprint fiber waste, whereas a slight increase can be noticed for the untreated waste. This increase may be due to the increase in the polar groups due to the increase in the effective dipole moment.

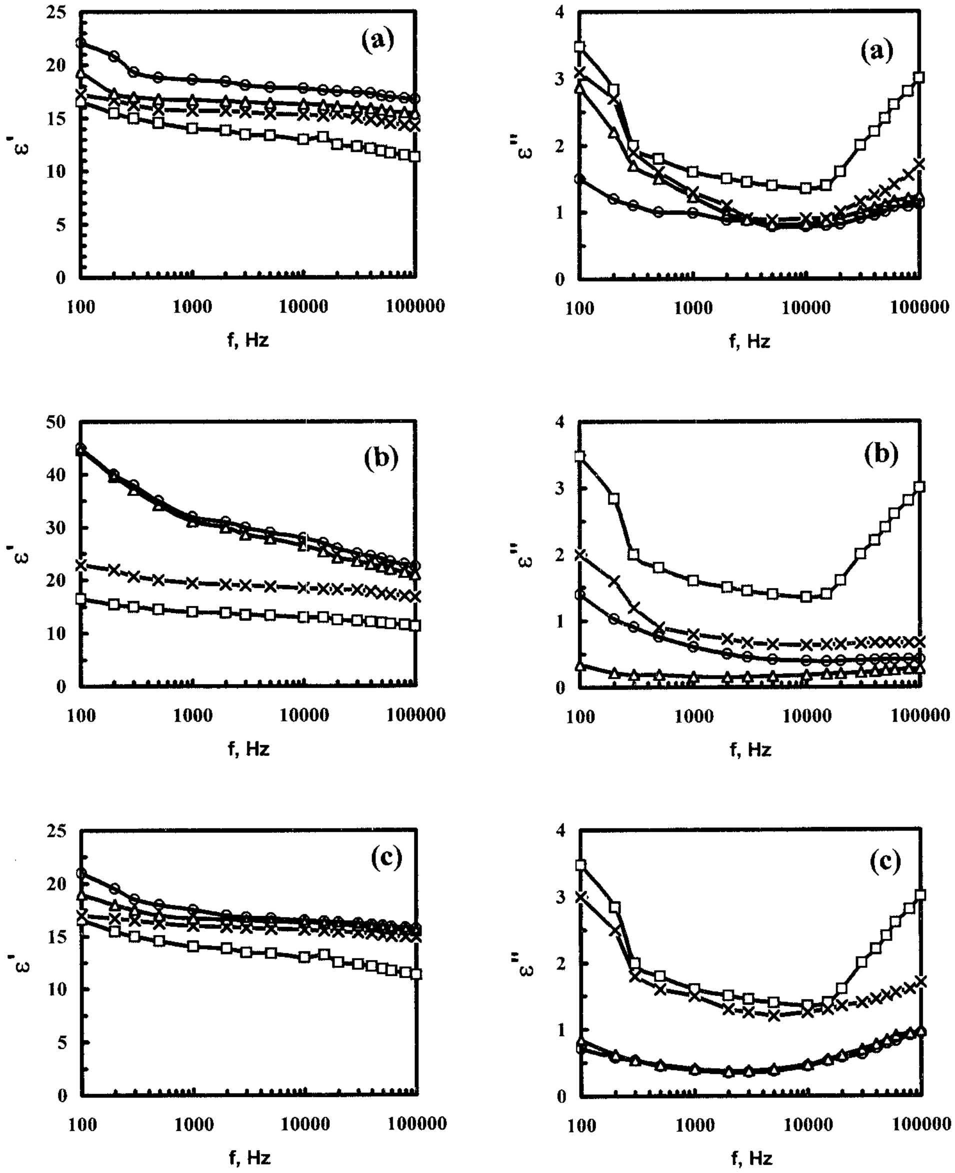

Figure 6 ϵ' and ϵ'' of NBR with the adhesion system (HRH) loaded with (a) untreated newsprint waste, (b) treated newsprint waste, and (c) silica at room temperature (25°C): (□) 0, (×) 20, (△) 40, and (○) 60 phr.

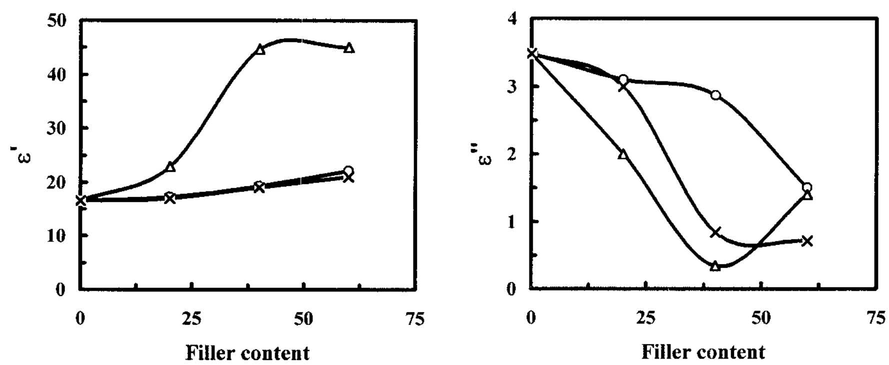

Figure 7 ϵ' and ϵ'' at 100 Hz of NBR with the adhesion system (HRH) loaded with different contents of (○) untreated newsprint waste, (△) treated newsprint waste, and (×) silica.

From this investigation, it can be concluded that the addition of both silica and untreated newsprint fiber waste increases ϵ', at a frequency of 100 Hz, from 16 to 21 and reduces ϵ'' from 3.5 to 1.4. However, at the same frequency, the addition of treated newsprint fiber waste increases ϵ' to 45 and reduces ϵ'' to 0.34 for a sample with a concentration of 40 phr. Therefore, the addition of treated newsprint fiber waste with a concentration of 40 phr could lead to an end product characterized by good electrical and mechanical properties.

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