



Modified Ground Rubber and its Effect on Physico-mechanical and Dielectric Properties of Nitrile-Butadiene Rubber Composites for a Green Environment



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Abstract

Around the world, the rubber industry uses 50% of primarily petroleum-based materials with the exception of natural rubber. The use of these petroleum-based raw materials results in increased environmental hazards in addition to the depletion of natural resources. The issue of ground rubber (waste tire rubber) is enormous and has broad environmental and financial consequences. There are several methods for recycling rubber, including devulcanization, grinding, surface treatment, and reclamation technology. This study aims to investigate the effect of ground rubber (GR) and their concentration on the rheological, morphological, mechanical, physical, and electrical properties of NBR composites. The impact of the chemical treatment ground rubber (GR) on the desired properties is also investigated. The investigated samples are prepared by using two roll mill mixer followed by compression molding at 162 °C. The SEM observations indicate some incompatibility and inhomogeneity in the blends, due to low interfacial adhesion between NBR and GR. However, by replacing GR by modified m-GR particles a more homogenous structure and uniform particle dispersion found, due to improved physical/interfacial interactions. The results also show that the rheological parameters, tensile strength and elongation at break are affected by the treatment of m-GR. The values of permittivity ϵ' improved after GR modification. Besides, the NBR composites containing m-GR have a promising insulating performance and can be used in electric insulation requirements. Additionally, the values of conductivity of the NBR/ GR and NBR/ m-GR composites are close to $\sim 10^{-6}$ S cm^{-1} at higher GR loading. This value is sufficient for these composites to exhibit electrostatic dissipation behavior.

Keywords: Nitrile Butadiene Rubber (NBR); Composites; Ground rubber (GR), Rheometric properties; Physico- mechanical properties; dielectric properties.

Introduction:

Due to its cross-linked structure and various additives, ground rubber (GR) does not disintegrate naturally for a very long time [1]. Additionally, they couldn't be disposed of in a landfill since the soil would absorb dangerous substances like oils. Utilizing waste tires as a raw material and recycling them would be the most advantageous strategy from an economic and environmental standpoint. Rubbers, and trace amounts of organic and inorganic fillers/additives are all common materials found in tires. The tire's primary component, carbon black,

accounts for 30-35% of its weight and offers a way to reclaim this carbon for use in energy-related applications. [1, 2].

Since there are a lot of rubber wastes generated nowadays and there are so many problems with the environment, recycling ground rubber has become an interesting subject for the scientists. The crosslinks produced during vulcanization are the main problem with rubber recycling. Rubbers become insoluble, thermally stable, and difficult to recycle after curing [3]. The development of science-based rubber recycling technologies and procedures

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as well as the usage of recycled rubbers in finished products will greatly reduce global energy consumption, provide renewable rubbers from rubber waste, and reduce environmental pollution. Recycling is one of the greatest solutions when taking into account the economic and environmental benefits. The use of waste/recycled rubber in polymer blends has commonly been carried out. These blends can be used to create a variety of products, including recycled rubber flooring, playground surfaces, and sports mats [4].

There have been numerous physical and chemical attempts to recycle the vulcanized rubber. Many physical and chemical attempts have been made to recycle the vulcanised rubber. Mechanical [5-7], thermomechanical [8-11], cryomechanical [9-10], microwave [10-11], and ultrasound [11] are examples of physical techniques. Chemical reclamation processes use various disulfide [12], thiol [13], and catalyst types.

Because the crosslinked GR particles and the polymer have weak interfacial adhesion, blending ground rubber (GR) with other polymers has produced poor properties [14]. To improve the adhesion between the two phases, waste rubber and polymer matrix, modification of waste rubber has therefore been carried out in the past [15].

Reclaimed rubber and polymer components can be combined with ground rubber (GR) in the form of ground particles. To improve the GR powder's adherence to rubber, the surface was modified. Through atmospheric pressure dielectric barrier discharge (DBD), the hydrophobic surface of GR powder has been changed to a hydrophilic one during the surface treatment procedure. Blends of surface-modified GR and NBR have enhanced mechanical characteristics, which are explained by an improvement interfacial bonding between the modified GR and NBR. Products made from NBR are employed in a variety of industries, including the automotive, building, aerospace, petroleum, and others. The most desired rubber is nitrile rubber (NBR), particularly for applications that are exposed to fuels, natural gas, and oil [3].

The addition of ground rubber (GR) has improved the tensile strength, modulus, and elongation at break of the composites as well as their impact resistance and thermal insulation. The soft phase of rubber gives the

composites flexibility. Based on these findings, it can be deduced that using rubber waste for a variety of successful and diverse uses has been demonstrated. Importantly, the creation of composites using rubber-based fillers helped to improve the environment [16].

It is aimed to study, how the amount of ground rubber (GR) affected the rheological, mechanical, and dielectric characteristics of nitrile-butadiene rubber composites. It was also stated how ground rubber (GR) modification affected the various examined parameters.

Materials

Nitrile rubber of acrylonitrile content of 32%, specific gravity 1.17 6 0.005 g/cm³; N-cyclohexyl-2-benzothiazole sulfonamide (CBS), pale gray powder, with a specific gravity of 1.27-1.31 g/cm³ at room temperature (25 -18 °C), melting point 95-100 °C was used as an accelerator. Zinc oxide and stearic acid were used as activators with a specific gravity at 158C of 5.55-5.61 g/cm³ and 0.90- 0.97 g/cm³ , respectively. Elemental sulfur is fine pale yellow powder, with a specific gravity of 2.04-2.06 g/cm³ at room temperature as used as a vulcanizing agent. Dioctyl phthalate (DOP) was used as a plasticizer with specific gravity 0.991 g/cm³ and boiling point 5 384oC. Polymerized 2,2,4-trimethyl 1,2-dihydro-quinoline (TMQ) was also added as an antioxidant. All the rubber ingredients were of commercial grades, purchased from Aldrich, Germany. Ground rubber powder (GR) was supplied by El NASR Company for rubber products (Tanta, Egypt).

Experimental procedure

Modification of GR; Synthesis of polyaniline (PANI/ GR) composite

For the preparation of modified ground rubber, equally weighted mixture of GR and pluronic acid was heated at 150 °C (oven) (see Fig. 1). After 4 h, turned into highly reclaimed state was taken out of the oven and waited until ambient conditions reached. After that an aniline monomer (0.1M) was dissolved in 50 ml methanol and placed with continuous stirring in an ice bath (0-5°C). A solution of (0.25 M) Ammonium persulfate (APS) as an initiator in 50 ml distilled water was rapidly added. The color rapidly darkened and after continuous stirring for 6 hours the reaction mixture was left overnight until having a

precipitate. Then precipitate is left drying in an electric oven under vacuum at 50°C. Polyaniline PANI is firmly prepared with GR and the PANI/GR swelling with green plasticizer composite formed. Finally the obtained modified (m-GR) was applied in different ratios (10, 20, 30, 40 phr) with NBR/HAF matrix and characterized.

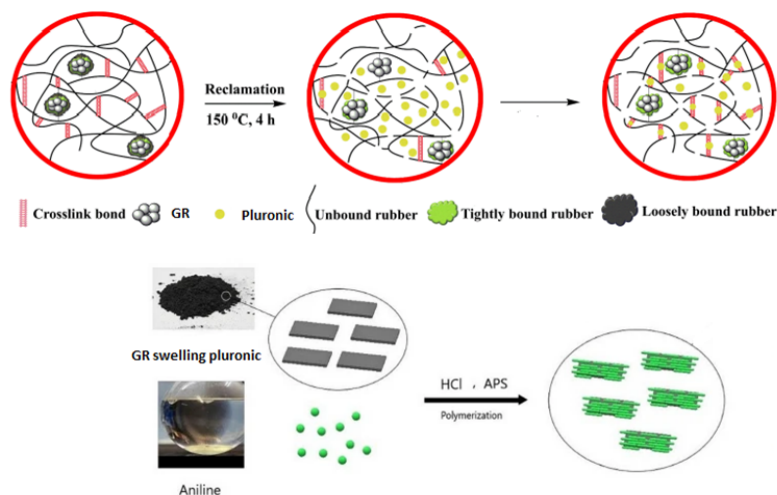


Fig.1. Schematic flow-chart of the preparation of modified ground rubber (m-GR)

Table (1): Formulation of the mixes in phr* samples

Ingredients	Samples								
	N0	N1	N2	N3	N4	N5	N6	N7	N8
NBR	100	100	100	100	100	100	100	100	100
ZnO	4	4	4	4	4	4	4	4	4
Stearic acid	2	2	2	2	2	2	2	2	2
HAF	25	25	25	25	25	25	25	25	25
GR	0	10	20	30	40	0	0	0	0
m-GR	0	0	0	0	0	10	20	30	40
DOP	2	2	2	2	2	2	2	2	2
TMTD	1	1	1	1	1	1	1	1	1
CBS	0.8	0.8	0.8	0.8	0.8	0.8	0.8	0.8	0.8
Sulfur	2	2	2	2	2	2	2	2	2
TMQ	1	1	1	1	1	1	1	1	1

* (Parts per hundred part of rubber)

Preparation of NBR/ waste rubber (GR) and NBR/modified waste rubber (m-GR) composites

Different formulations were used in the preparation of NBR/ ground rubber (GR) and NBR/modified ground rubber (m-GR) composites as shown in Table (1). Different loadings of GR and m-GR were used from 0 to 40 phr (i.e., parts of hundred part of

rubber). A laboratory two-roll mill was used to mix the ingredients at room temperature with outside diameter of 470 mm, working distance = 300 mm, speed of slow roll = 24 rpm and fraction ratio of 1: 1.4. After mixing, the rubber compound was conditioned at 25°C for 24 h prior to cure assessment.

Curing characteristics

Using a Monsanto Rheometer (Tech-Pro, Cuyahoga Falls, OH, USA), the curing characteristics of the mixture were examined in accordance with ASTM method D2084-11. The samples in concern were tested at a vulcanization temperature of 162 + 1°C. The rheograph was used to calculate the scorch time (measured as t_{s2}) and the cure time (measured as t_{c90}).

Vulcanization process

A compression molding machine (Mackey Bowley, C1136199) was used in each instance to cure the compounded rubber stock for the appropriate optimal cure time (t_{c90}), which were determined using rheometric properties. Before testing, the vulcanized sheet was kept for 24 hours in a cold, dark place after curing.

Instrumentation

Fourier transforms infrared spectroscopy (FTIR):

A JASCO FT-IR-6100 was used to record (FTIR) spectra. Transmission mode tests were conducted on the samples between 4000-400 cm^{-1} .

Mechanical properties:

According to ASTM D412-15a, tensile strength (MPa) and elongation at break (%) were determined for the composites using Zwick Z010 testing equipment (Germany). Dumbbell-shaped samples were created by cutting the molded sheets with a Wallace die cutter at a crosshead speed of 500 mm/min. The experiments were conducted at room temperature.

Scanning electron microscopy (SEM):

The SEM images of NBR/ground rubber (GR) and NBR/modified waste rubber (m-GR) composites were acquired using a Leica Cambridge S-360 SEM, a model made in Wetzlar, Germany. Prior to observation, all the fracture surfaces for the chosen samples were collected by breaking the specimens under liquid nitrogen.

Dielectric properties:

At room temperature $\sim 30^\circ\text{C}$ over a broad frequency ranging from 0.1 Hz to 1 MHz, the permittivity ϵ' , dielectric loss ϵ'' ; ($\epsilon'' = \epsilon' \cdot \tan\delta$, $\tan\delta$; is the loss factor) and alternating resistance R_{ac} were obtained. An impedance analyzer was connected to a personal computer through a GPIB cable (IEE488) to computerize the measurement. Data collection was using the commercial interfacing and automation software application "Lab VIEW". There is an error in ϵ' and $\tan\delta$ amounts to $\pm 1\%$ and $\pm 3\%$, respectively. A temperature regulator with a (Pt 100) sensor was used to control the samples' temperature. The measurement error for temperature is $\pm 0.5^\circ\text{C}$.

Results and discussion:

The FTIR spectra:

The modified GR's FTIR spectra (Fig. 2) show that as peak intensity increased, all peaks became more pronounced. The surface modification had led to the GR changing and becoming more hydrophilic. Higher polarity increases the surface energy of the rubber and is anticipated to start more linkages with the polar groups of the NBR, increasing the composite's tensile strength and improving adhesion between the GR as filler and the NBR, consequently increasing composite strength.

The well-known technique for enhancing rubber-matrix interfacial adhesion is surface treatment. However, because they migrate to the surface of rubber products and create a passive weak layer, the additives can contribute to poor adhesion between the rubber and the GR. In addition, the highly cross-linked rubber structure brought on by the vulcanization process may contribute to poor matrix adhesion [17].

The similar adsorption peaks in the FT-IR spectra of GR and modified m-GR show that the two compounds share many chemical characteristics. On the original and modified carbon, there are absorption peaks at 3400 cm^{-1} that are caused by the -OH vibrations connected to hydrogen bonding, phenolic hydroxyl, and N-H stretching vibrations. The intensity of the -OH peak is significantly increased between 3000 and 3500 cm^{-1} , and a broad band is seen that is attributed to the hydroxyl groups formed as a result of oxidation, indicating that the modification has increased the number of -OH (Fig. 2 (a)). A new signal that has carbonyl group origins can be seen at 1650 cm^{-1} . As a result of saturated primary alcohols' C-O vibration, bands at 1081 and 1013 cm^{-1} are seen to appear. N-H stretching vibrations and C-O stretching vibrations, respectively, can be attributed to the bands at 1726 cm^{-1} and 1013 cm^{-1} (Fig. 2 (b)).

The NBR with m-GR spectrum appears to be a superposition of the spectra of the two components, with NBR absorption bands predominating. In particular, absorptions at 1140 cm^{-1} , 1490 cm^{-1} , and 1585 cm^{-1} have been attributed to amine C-N stretching, C=C stretching of the aromatic rings, and C=N stretching, respectively.

These findings will encourage increased efforts to recycle waste rubber (GR) in order to address

environmental issues related to large amounts of waste rubber.

Rheological Characteristics for NBR/ 334 ground rubber (GR) composites:

The results are tabulated in Table (2) for the rheometric properties of the rubber compounds expressed in terms of the minimum torque (ML), maximum torque (MH), and ideal cure time (Tc90), scorch time (ts2), and cure rate index (CRI).

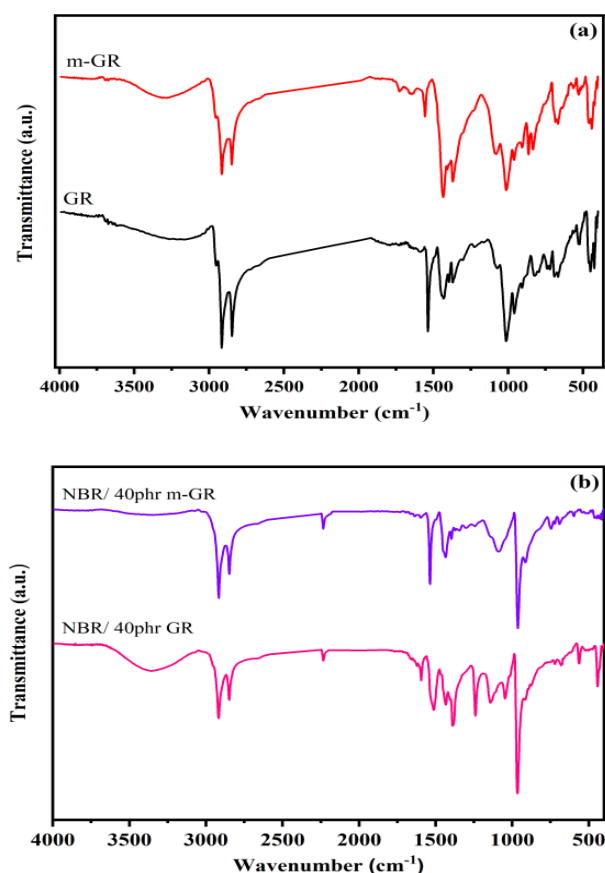


Fig. 2. (a) The FTIR spectra of the GR and the modified m-GR. (b) NBR/ GR composites and NBR/ 40 phr m -GR composites.

The viscosity of the compounds, which increased with the increase in GR, is related to the minimum torque, or torque at the initial stage of vulcanization. Maximum torque (MH) is dependent on the degree of crosslinking as well as the reinforcement provided by the polymer matrix's particles. The values of (MH) in

Table 2 decreased with the increase of GR. This is because the rubber composite has become more polar. The decreasing torque differential between NBR & GR suggests that cross-linking was not

distributed evenly, leading to over cross linked and poorly cross-linked phase, which lowers the

Table (2): Rheological characteristics of the NBR/ ground rubber (GR) composites.

Sample	M _L (dN.m)	M _H (dN.m)	t _{c90} (min)	t _{s2} (min)	CRI (min ⁻¹)
N0	0.97	19.84	5.71	2.24	28.8
N1	1.14	19.47	5.38	2.22	31.65
N2	1.29	19.34	5.08	2.14	34.01
N3	1.49	19.04	4.90	2.11	35.84
N4	1.65	18.14	4.79	2.10	37.17

M_H value. It is observed that the CRI rises with the addition of GR. Poor interfacial interaction between NBR and GR was observed at higher loadings of GR, confirming the filled compounds' propensity to form filler agglomerates, as represent in table 2 t_{c90} decrease as show in table 2 [18, 19].

Mechanical properties of NBR/GR composites

In the polar host matrix NBR/ GR composites, the impact of waste rubber loading on tensile strength and elongation at break is presented in Fig. 3 (a,b). It is evident that the tensile strength decreased by up to 20 phr then a slight increase encountered at 30phr but still lower than the value of the blank sample. This decrease principally upon increasing the concentration of GR can also be attributed to the poor dispersion of GR into NBR matrix, which is illustrated later in the SEM section. Elongation at break values exhibit the same behaviour.

Morphology of NBR/GR composites

The SEM photographs are shown in Fig. 4. Using SEM, the fractured surfaces of the NBR/GR composites were examined and contrasted with those of the control sample Fig. 4, a.

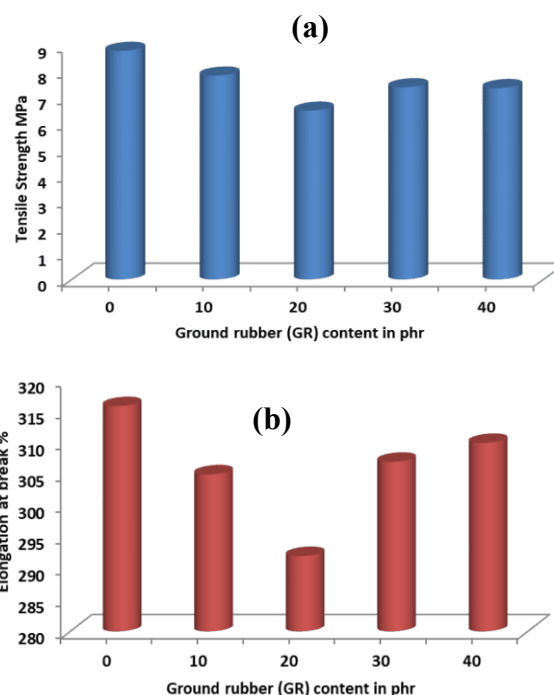


Fig. 3 (a, b). Tensile strength and elongation at break of NBR/GR composites are dependent on the amount of ground rubber (GR).

A significant number of agglomerates were clearly seen, which suggested that NBR and GR had poor interfacial adhesion. There was more of a lack of adhesion in 30, 40 phr of GR than that in 10 phr concentration of GR because the GR particles agglomerates have started to form after 10 phr Fig. 4, b. However, the weak adhesion damages or modifies

the mechanical properties, which is consistent with the interpretation of the experimental data (Fig. 3).

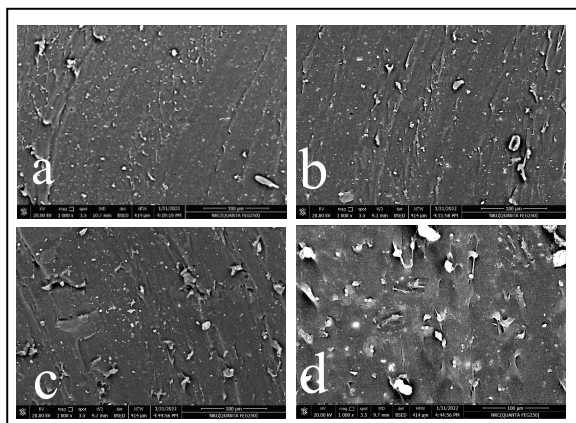


Fig. 4: SEM micrographs of NBR/GR composites; (a). NBR/0phr GR (b). NBR/10phr GR, (c). NBR/30 phr GR, (d). NBR/40 phr GR.

Rheometric characteristics for NBR/ modified ground rubber (m-GR) composites:

GR was chemically modified by using polyaniline (PANI) to improve the dispersion of GR into NBR matrix, where m-GR interfacial strength is crucial to achieving good filler reinforcement [20, 21]. From, Table 3, the value of MH increased upon increasing m-GR, indicating enhancement of crosslinking density. The strong interphase adhesion between NBR and m-GR up to 30 phr could be used to demonstrate the increase in crosslinking density. Agglomeration of the m-GR in the NBR matrix, as previously observed by SEM (Fig. 4, c), caused the maximum torque to decrease as the m-GR loading was increased further (to 40 phr). The optimum cure time, t_{c90} (time for 90% cure), as well as the scorch time, t_{s2} (the initial vulcanization time) increased due to the presence of m-GR, indicating that the vulcanization reaction of NBR was accelerated. With the addition of m-GR, it is observed that CRI rises, supporting the activation of NBR's cure reaction [16, 18]. According to prior research, this could be explained by the reactive functional groups in m-GR acting as activators and speeding up the rate of

vulcanization. With the addition of m-GR, CRI is seen to rise as shown in Table 3, supporting the activation of NBR's cure reaction up to 30 phr of m-GR before declining [16,18]

Table (3): Rheological properties for the composites of NBR and m-GR

Sample	M_L (dN.m)	M_H (dN.m)	t_{c90} (min)	t_{s2} (min)	CRI (min^{-1})
N0	0.97	19.84	5.71	2.24	28.8
N5	0.96	20.67	6.14	3.99	46.51
N6	0.88	23.47	6.68	4.83	54.05
N7	0.85	24.46	8.21	6.89	75.76
N8	0.83	24.11	8.19	6.75	69.44

Mechanical properties of NBR/ m-GR composites

The effect of modified ground rubber (m-GR) loading on the tensile strength and elongation at rupture for the polar host matrix NBR is presented in Fig. 5 (a,b). The tensile strength of these compounds clearly increased up to 30 phr before significantly decreasing at 40 phr, the maximum loading concentration is increased, and the adhesion system provides the best mechanical properties up to 30 phr. This increasing principally upon loading m-GR can be also in relation to the even dispersion in the rubber matrix (shown later in the SEM section). However, the values of elongation at break values exhibits the same behaviour. However, the decreasing in the mechanical parameters at 40phr was attributed to the anticipated m-GR agglomeration at high level (beyond 40 phr), which was no longer adequately separated or wetted by rubber phase, making the stress transmission from matrix to m-GR difficult [20].

SEM of NBR/ m-GR composites

The micrographs of the NBR composites containing modified ground rubber (m-GR) are shown

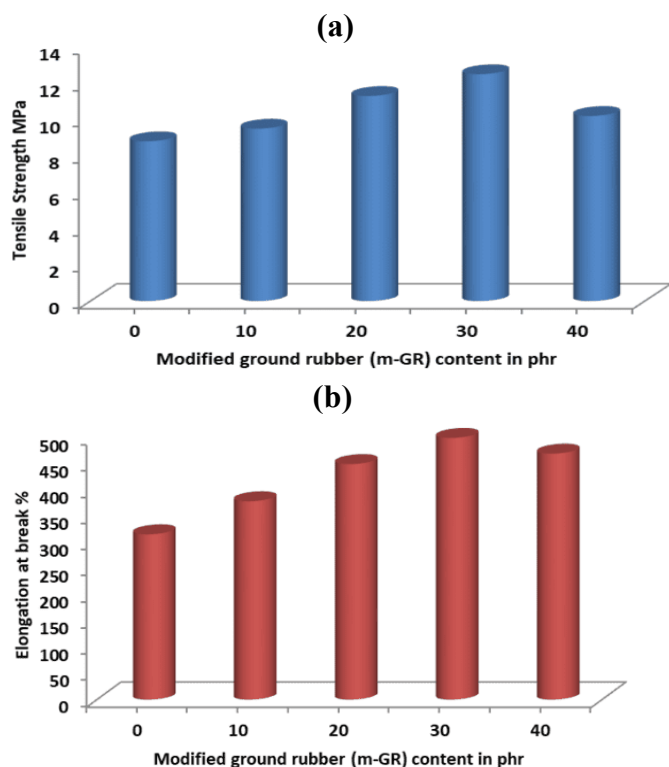


Fig. 5. Tensile strength and elongation at break of NBR composites are dependent on the amount of modified ground rubber (m-GR).

in Fig. 6, there was a noticeable difference in the morphologies and the unmodified NBR composites (Fig. 4). However, in case of the modified NBR/m-GR composites. In the NBR matrix, the SEM images revealed a finer and even better dispersion of m-GR up to 30phr (Fig. 6, b). In contrast, in Fig.6, c. In contrast, in Fig.6c. There were some interfacial adhesion issues between NBR" and m-GR, due to higher m-GR concentration (40phr), m-GR particles began to form agglomerates as they were no longer effectively moistened by the rubber phase. Further, the dispersion of m-GR 40 phr in NBR/ m-GR material (Fig.6, c) seems better compared to that of NBR/40phr GR (Fig.4, d) without modification. The Morphology studies of the NBR/m-GR composites were in excellent agreement

with the variation in the mechanical parameters in Fig. 5.

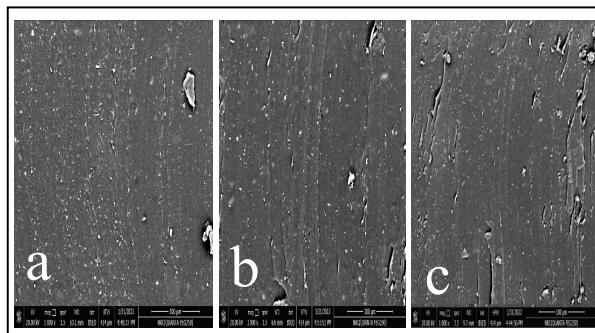


Fig. 6. SEM micrographs of NBR composites containing different m-GR loading; (a). NBR/10phr, (b). NBR/30phr, (c). NBR/40 phr.

Dielectric characteristics of NBR/GR and NBR/m-GR composites:

Figs. 7 (a) and 8 (a), display the permittivity ϵ' and dielectric loss ϵ'' data for NBR/GR and NBR/m-GR composites, when plotted against the applied frequency f . Since the dipole can no longer follow the presented alternating field at higher frequencies, the values of ϵ' clearly decrease with frequency and reach a steady value there. Additionally, NBR/GR composites have higher permittivity at lower frequencies because the majority of the free dipolar functional groups can orient at these frequencies. Further raising the frequency of the applied voltage makes it more challenging for large dipole groups to orient, which causes permittivity to decrease as frequency rises. However, the values of ϵ' improved after GR modification, as depicted in Fig.8 (a).

In contrast, the dielectric loss ϵ'' in Fig. 7(a) and Fig 8(a) decreases as the frequency increases and increases with the increase of GR and m-GR loading. The frequent increase in conductivity and interfacial polarization are the main causes of the increase in dielectric loss. Interfacial polarization takes place in heterogeneous systems, such as multi-component.

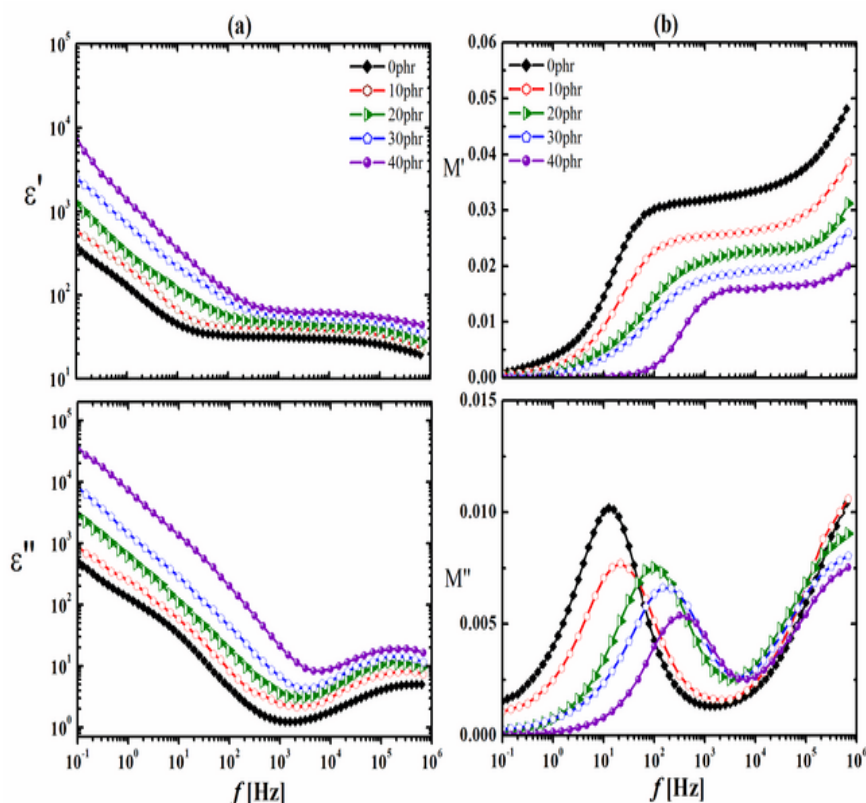


Fig.7. (a). Permittivity ϵ' and dielectric loss ϵ'' for NBR/GR composites are plotted against frequency f .
 (b). dielectric modulus (real M' and imaginary M'') as a function of frequency f at (~ 30 °C).

systems, when accelerated charge carriers are captured and hindered by the physical barriers. Additionally, this makes the system more polarised. The conductivity contribution, which is more evident in the lower frequencies, may be also the cause of the larger values of ϵ'' . However, the curves ϵ'' are broad which suggests the existence of several relaxing mechanisms. The complex electric modulus M^* ($M^* = 1/\epsilon^*$) can be used to explain the relaxation mechanism of the investigated systems [22- 24].

Certainly, the information in the electric modulus and permittivity spectra is identical. But in the frequency dependence of M^* , a very obvious peak frequently and generally arises. In Figs. 7b and 8b, two relaxing processes can be found. The Maxwell-Wagner effect or interfacial polarisation may be responsible for the first relaxation. [25, 26], which operates in the

lower frequency range. The absorption in the higher frequency region may be caused by the segmental orientation of the polymer chains, which can also be found elsewhere [25, 27].

However, Fig. 9 summarizes the dielectric results; Loss factor $\tan \delta$ and permittivity ϵ' ($\tan \delta = \epsilon''/\epsilon'$) at stationary frequency (100 Hz) for comparison. Obviously, the values of ϵ' improved after GR modification. The values of $\tan \delta$ are decreased. This means that the NBR composites containing m-GR have a promising insulating performance and can be used in electric insulation requirements [28].

Electrical conductivity of NBR/GR composites

Figures 10 a,b show how the conductivity of NBR/GR composites varies with frequency before

and after GR modification, respectively. As can be seen, the conductivity increases as frequency rises;

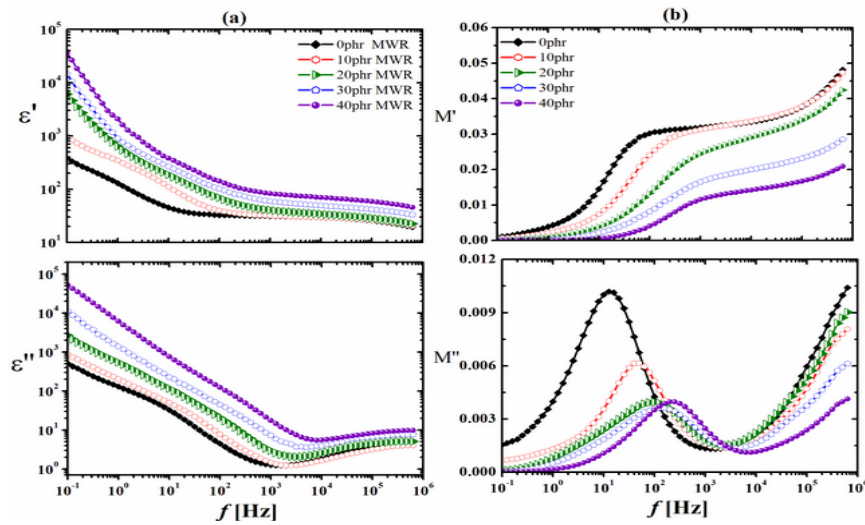


Fig. 8. (a). Permittivity ϵ' and dielectric loss ϵ'' for NBR/m-GR composites are plotted against frequency f . (b). The dielectric modulus (real M' and imaginary M'') as a function of frequency f at room temperature ($\sim 30^\circ\text{C}$)

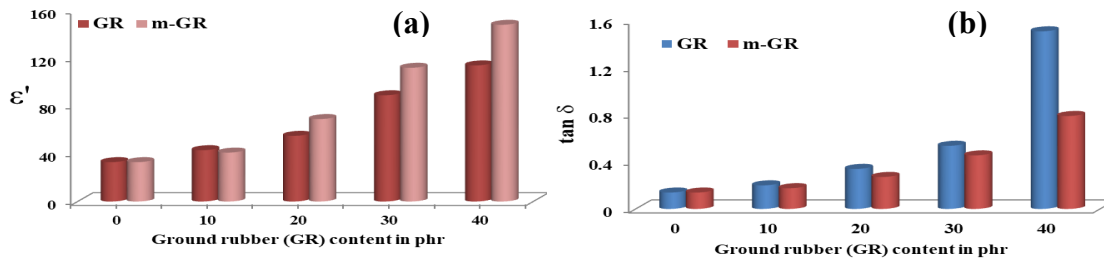


Fig. 9 (a,b). The permittivity ϵ' and the loss factor $\tan \delta$ vs. GR concentration (phr) at fixed frequency 100 Hz before and after GR modification .

it reaches a plateau at low frequency as GR content rises. However, the GR agglomerates in the matrix are responsible for the significant increase in conductivity. Additionally, at higher GR loading, the conductivity values of the NBR/ GR and NBR/ m-GR composites are close to $\sim 10^{-6} \text{ S cm}^{-1}$.

The electrostatic dissipation behaviour of these composites can be attained with this value [29].

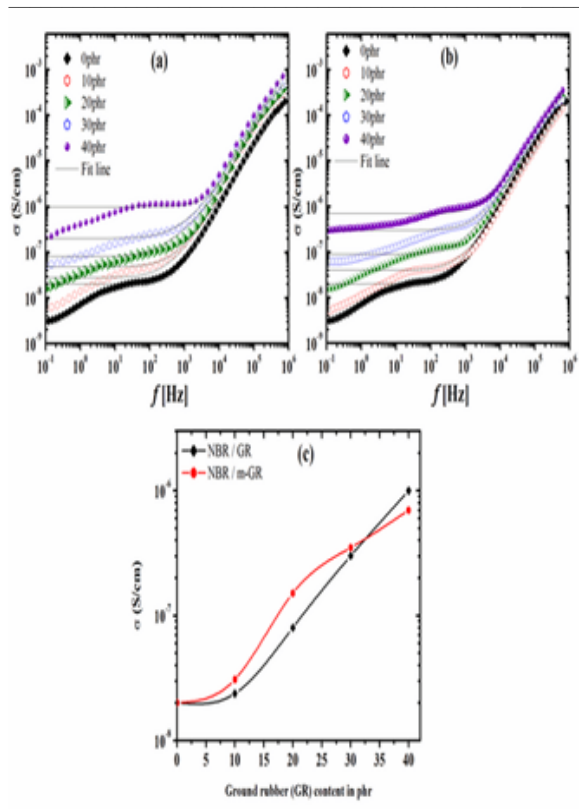


Fig. 10 (a,b). The electrical conductivity σ vs. frequency f for NBR/GR before and after GR modification. (c). Dependence of conductivity σ on GR and m-GR content.

Additional, the conductivity follows a power law for every proportion being studied [29, 30] Eq. (1);

$$\sigma = \sigma_{dc} + \omega^s \quad (1)$$

where σ_{dc} is DC conductivity, ω is the angular frequency, A is a frequency independent parameter and s is a power, the values of s are found to be $s \leq 1$, which is governed by electronic hopping processes.

Moreover the values of DC conductivity σ_{dc} obtained from the fitting process are plotted versus GR content as shown in Fig. 10 c. As the GR content is raised a sharp increment in conductivity is noticed with increasing GR ratio.

Conclusion

In the current investigation, two roll mill mixers were used to prepare several NBR

compounded rubber comprising various ratios of ground rubber (GR) up to 40 phr (prior to and following GR modification). Strength and elongation at rupture improved by GR modification up to 30 phr. Low interfacial adhesion between NBR and GR is revealed by the SEM investigations. Uniform particle dispersion was noticed as m-GR particles were added. Permittivity (ϵ') values are increased by GR modification. Additionally, the NBR composites containing m-GR show promising insulation properties and can be employed for electrical insulation needs. For greater GR loading, the conductivity values of the NBR/ GR and NBR/ m-GR composites are not far from 10^{-6} S cm^{-1} . The electrostatic dissipation behaviour of these composites can be achieved with this amount. However, the values of σ enhanced by GR modification up to 30 phr m-GR.

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Conflicts of interest: Authors declare that there no Conflicts of interest.

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