

The Influence of Graphene Oxide Addition on The Fortified Nitrile Butadiene Rubber Nano-composite Qualities

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Abstract

Strengthening elastomers by the Nano-fillers, for example, Graphite subordinates, have high embellishments on their attributes. In the present work, Graphene Oxide (GO) nano-sheets have scattered in huge rubbers was Nitrile Butadiene Rubber (NBR). Enhancement the vulcanization procedure on (GO/NBR) nano-composite elastic was performed by arrangement blending strategy and rumination blending method. Scattering of GO into NBR was homogenous, where affirmed by Scanning Electron Microscopy (SEM) and X-beam Diffraction (XRD) test. Breaking down the useful gatherings by Fourier-Transformed Infrared Spectrophotometry (FTIR) has been explored as well. Diverse stacking wt% of GO inside NBR has clear impacts on the rigidity and flexibility conduct of the nano-composite. Impact of GO on the electrical conductivity and persuasive properties of GO/NBR nano-composite elastic portrayed too. The GO presence has upgraded the crosslinking trademark in GO/NBR nano-composite elastic and enhanced its properties.

Keywords: Graphene oxide (GO), Nitrite Butadiene Rubber (NBR), Dispersion (GO) in the elastomers, (GO/NBR) nano-composite rubber.

Introduction

The enforcement and improvement of various plastics won a lot of attention lately, and has received considerable attention in the sectors of industry and scientific research. Synthetic rubber with high geometric properties is used in many applications, but it is not self-reinforced rubber [1]. However, strengthen it must be done by adding special fillers usually effective as an agent to improve the tensile strength and oil resistance, as well, as the thermal stability of the rubber [2, 3]. The addition of nanoparticles (1-100 nm) to the polymer improves its specifications by changing many properties in it. [4]. Fillers are divided into two main types in the rubber industry, black and non-black fillers. The purpose

of these fillers is to improve the mechanical, thermal, and environmental effects. [5].

The nitrile butadiene rubber (NBR) is one of the main types of plastics, which consists of acrylonitrile ($\text{CH}_2 = \text{CHCN}$) and butadiene ($\text{CH}_2 = \text{CH}-\text{CH} = \text{CH}_2$). The single molecule of the two interrelates with other molecules to form multi-unit large molecules through the bonding between free radicals. (NBR) has the structure of a series of irregular and amorphous making it unable to be extended the crystallization [6]. NBR is used in a wide range of applications such as fuel and oil hoses, and seals [7]. NBR becomes attractive when the compound enhances by the addition of graphite derivatives (where carbon is the backbone of her). Graphene / NBR compound requires a great care in the selection of the type of used graphene nanoparticles, its matrix, its manufacturing and producing process, and defining its specifications [8].

The main challenge in the function of graphene polymers is the poor compatibility between the graphene and the indigenous polymer molecules. However, there are lots oxygen groups link to the graphene surface which are connected to the Graphene oxide (GO). This oxygen groups allow the dispersal of the GO in aqueous solution in proper form. It also reduces the interaction between the host polymer and GO through covalent or non-covalent bonds [9]. Where, GO is a monolayer material is formed by the oxidation of graphite followed by exfoliation treatment by ultra-sonication technique [1]. GO has similar mechanical properties of carbon nanotubes (CNTs); because it has excellent electrical and thermal properties because of the large area, which reaches up to about $2600 \text{ m}^2/\text{g}$, compared with $1350 \text{ m}^2/\text{g}$ of carbon nanotubes (CNTs) [4]. The interface between the layers of GO nano-sheets when deployed in the rubber matrix play an active role in the re-creation of the structure of the rubber-nano composite and determine its properties [7].

It is very important and necessary for the production of GO / plastics nano-composites the careful in taking advantage of the benefits of enhancement GO properties through the use of

several different production techniques [10, 11]. It was found that the preparation of nano compounds-vulcanizes NBR and the integration of GO has been tested by a number of research groups such as Li et al. [12], and who explored GO effect on the mechanical properties of the NBR. Bay and others [13] found that the glass transition temperature (T_g), wear resistance, and tensile strength of the GO / carboxylated hydrogenated acrylonitrile butadiene rubber (HxNBR) strength has been improved. Homogeneous dispersion of GO in the rubber matrix confirmed through changes in the interlayer distance that were calculated from XRD data [14].

The polymer becomes a cross-linking when subjected to some organic solvents, where it absorbs part of the solvent and thus amplified instead of completely dissolves. The ingestion of the solvent force the polymer chains into the polymer network to elongate, which leads to the generation of re-tractive force in the area of occurrence of this distortion [15]. The cross-linking process plays an important role in determining the cause of physical properties of vulcanization [16].

The purpose of the current work is to prepare GO / NBR nano- composite rubber by adding GO to NBR matrix in water base and solvent compatible. GO vulcanized / NBR rubber composite is produced using a poly drive internal mixer and two roll mill techniques. The produced material has been subjected to many tests to evaluate its properties and assessments. The effects of the GO addition on the crosslinking density in the GO / NBR nano-composite rubber is assessed, in addition to other parameters such as: the relevant functional groups and GO nano-sheets, and the affected mechanical properties of the NBR.

Material and methods

The nano-composite preparation

In this study, the NBR Krynac 3345 with the Acrylonitrile substance was 33 ± 1.0 wt.% and Mooney thickness ML(1+4), and Graphite drops < 20nm incorporates $KMnO_4$, concentrated H_2SO_4 , HCl were utilized. Adjacent to, numerous different materials, for example, Acetone, dimethylfuran (DMF), Zinc Oxide, sulfur, stearic corrosive, were utilized as a part of the trials to set up the required NBR-nano composite material.

Graphene Oxide (GO) manufacturing

GO was set up by utilizing the oxidizing graphite drop characteristic and changing the Hammer's strategy [17]. The procedure in a word, 400 ml of H_2SO_4 in a cup (1liter size) submerged

in a chilly water shower. Graphite (3g) was added to the carafe under blending utilizing attractive stirrer. At that point, 15g of $KMnO_4$ was gradually added to the above blend, the acquired material gets to be dark brown after around 60 hrs. Hydrogen peroxide (20 ml) was blended with 400 ml of refined water, and added to the arrangement gradually, with mixing process. The arrangement shading was changed from yellow to brown. At that point, the came about arrangement was washed by centrifugation process went on for 20 minutes at 9000 RPM. The washing agent was a solution of 5% of HCl and refined water. The washing procedure was reshaped three times to affirm the material virtue. The blend was washed again three times by deionized water and refined water. The procedure was done to affirm that pH of the blend was impartial. At long last, the GO slurry was decontaminated and dried in a vacuum stove at $60^\circ C$.

The preparation of GO/NBR nano-composite

The GO/NBR nano-composite rubber was carried out effectively by arrangement blending strategy as it is the less demanding and ideal methods to scatter GO into the NBR homogeneously. Finding the perfect and reasonable solvents was viewed as a crucial target in this study. Acetone 500ml was utilized to loose 10g of little sections of the elastic (NBR) to get a low consistency lattice. The required weight of GO arrangement arranged by peeling and scattering measure of GO in DMF by sonication process. The sonication procedure lasted for 50 min with a setting of 500 watt, 25 kHz, 20% adequacy at $30^\circ C$ of the ultrasonic vibrator, as Fig.1a appears. The GO/DMF suspension was continuously filled the NBR arrangement depicted already and blended by a mechanical stirrer for 2 hours as Fig.1b uncovers. Step by step, Deionized water was added to the composite blend to stay away from the development of sudden coagulation stage.

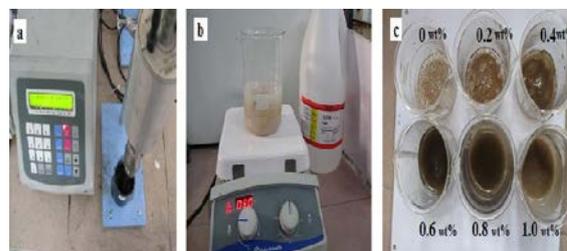


Figure 1: GO/NBR nano-composite rubber preparation; (a) - GO sonication, (b) - NBR dissolution, (c) – GO/NBR mixing and drying with different loading of GO.

The nanocomposite blend dried in a vacuum stove

at 65°C until its weight was unaltered as Fig.1c illustrates. The agents and additives were added to the dried pattern to cure it before the blended GO/NBR was placed in the two-move mills and afterward by a poly drive blender at the room temperature for 15 min. The compound definition and added substances recorded in the Table 1 communicated as part for every hundred sections of the rubber (phr). Different GO weight fractions (wt%) were utilized to provide GO/NBR nano-composites tests, which were dealt with in a pass on hot plates of (100x100x1) mm. The examples were squeezed to 10MPa in a squeezing machine in light of the ASTM D3182 at 130°C. Variable preheating times, squeezing with warming took after by cooling were completed by utilizing rheometer investigation information.

Table1: GO/NBR nano composite composition of different loading of GO wt%

Vulcanization	Ingredients (Phr)*
GO	Wt. %
NBR	100
Sulfur	2.00
ZnO	4.00
S. Acid	1.50
CBS(cz)	0.50
A-Oxidant	1.00

*phr, parts per hundred of rubber; CBS (CZ), N-cyclohexyl-2-benzothiazolysulfenamide;

Measuring instruments

X-ray diffraction (XRD)

The estimations of X ray diffraction (XRD) were completed to describe the structure of graphene oxide and NBR. An APD 2000 diffract meter in Bragg-Brentano geometry with monochromatic Cu K-alpha radiation (40 kV, 100 mA, k50.154 nm) was used . The XRD measures were obtained at 25°C in view of nonstop scan steps. The information gathered in the θ -2 θ mode.

FTIR analysis

Fourier-transform Infrared Spectroscopy (FTIR) sort Nicolet 6700 was utilized over a recurrence scope of 500-4000 cm-1. The test was done to portray the useful groups in the GO/NBR nano-composite elastic.

SEM test

Fig. 2 shows the surface micrographs of delegate tests of GO, NBR, and GO/NBR nano-composite utilizing the scanning electron microscopy (SEM) test. The GO sheets in (Fig. 2a) show up in layers of the network form. The surface way of NBR appears by means of the SEM examining likes one stage and state of plastic structure as in (Fig. 2b) and any distinction may show up due to the non-crystallite of the elastic districts.

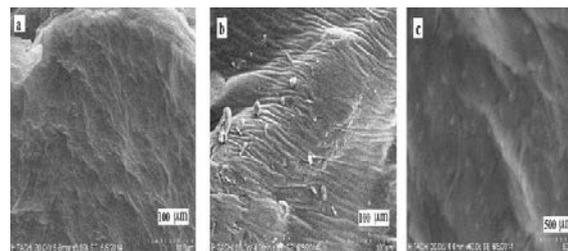


Figure 2; scanning emission microscopic test (SEM) of a- GO, b- NBR and c- GO/NBR

Fig. 2c shows adequate scattering of the GO sheets into the NBR framework, despite the fact that the scattering of GO in the elastic network was not clear. This scattering at 500nm scale was with few GO agglomerations; however, it here is a solid connection between GO sheets and the framework of NBR.

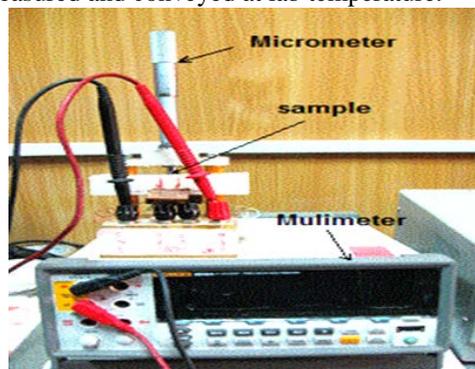
Tensile test

The tensile tests directed for GO/NBR nano-composite rubber specimens by utilizing an Instron machine (5566 10KN) as indicated by ASTM D412 benchmarks. The adequacy of variable GO loadings and its presence into the NBR grid was considered by watching the estimations of stress, strain and modulus. Notwithstanding the yield and breakpoints in light of the fractions of scattered GO in NBR was examined.

The electrical properties

Electrical conductivity was measured to confirm the GO nano-sheets scattering in GO/NBR nano-composite. Keithley-2000 multimeter was employed with an advanced voltmeter associated with two square steel plate of (20x20) mm², as (two focuses' props) cathodes associated with a micrometer as Fig. 3a manifests.

The dielectric consistent was measured by RF Impedance/Material Analyzer sort (Agilent 4291B) as appeared in (Fig. 3b). The frequency range was from 1MHz to 1.8 GHz, distinctive GO fractions in the GO/NBR nano-composite were measured and conveyed at lab temperature.



(a)

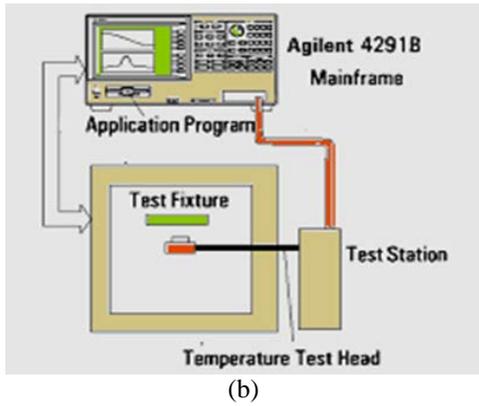


Figure 3: Electrical properties testing; (a) two points conductivity, (b) dielectric property

Results and discussions

Figure 4A illustrates the scanning electron microscope (SEM) picture, the results demonstrates the minute surface of the specimen stacked with 1wt% of GO. The specimen resembles a smooth example without introduction to strains and the GO scattering in the elastic framework was not clear. At the point when the specimen is extended by the elastic test, the SEM examination of the presence of an irregularity and wrinkly part leaves of the GO and the rubber lattice. Subsequently, this outcome gives a sign of the conceivable more grounded collaboration between the GO sheets and rubber matrix. The X-beam diffraction test affirmed this outcome. The keeping up a rehashed multi-layer structure in good figure in the intercalated structure made of divergences interfaces between sheets satisfactory for determination. The polymer chains intercalation, similar to the instance of the elastic and between the fillers layers expanded the dispersing between the layers, which prompts the crest X-beam diffraction points towards the low transformation.

Fig. 4b demonstrates of the FTIR spectra tests, the expansive crest of GO around 3282 cm⁻¹, it can be viewed as the consequence of hydroxyl O-H gathering and vibration perceived O-H extending. The crest of C=O of carboxylic corrosive at 2100 cm⁻¹, and C=C from un-oxidized GO surface at 1995 cm⁻¹. The range of perfect NBR indicates trademark retention crests at 3322cm⁻¹ (- OH), 2800-2900cm⁻¹ (- CH₂-), 2250cm⁻¹ (- CN). The NBR and GO/NBR nano-composite elastic shows comparative assimilation crests in their FTIR spectra. The crests that show up around 1400-1800 cm⁻¹ speak to the carbonyl extending. At 2100cm⁻¹, the retention expanded in view of (- C=O) of the carboxyl gathering in the GO/NBR nanocomposite containing 1wt% of GO. This is because of the arrangement of (- H) holding between the carboxylic gathering (-

COOH) of NBR and hydroxyl bunches (- OH) of the GO sheets.

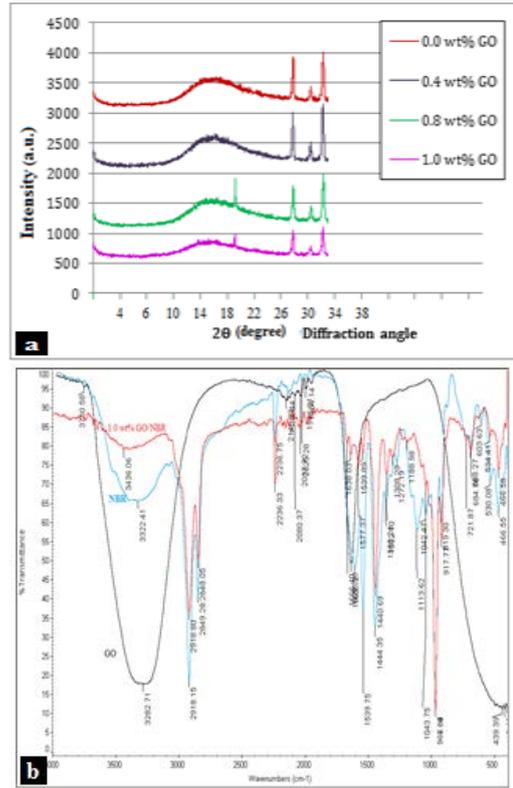


Figure 4; (a) X-ray diffraction test and (b) FTIR spectra tests for NBR and different loading of GO in GO/NBR nano-composites.

Fig. 5 demonstrates the conduct of the vulcanized GO/NBR nanocomposite rubber in stress-strain tests. There were straight changes in the stress-strain against GO fraction. The Modulus expanded as GO fraction expanded as well, where it expanded more than 200% for 0.8 and 1 wt% contrasted with the neat NBR. At 0.2-0.6 substance of GO wt%, there were no critical changes in the elasticity. There is an impact of GO fraction on strength quality and elasticity modulus, despite the fact that some anomaly in modulus due to the elastomer mechanical conducts and amorphous structure. The upgrade in the mechanical properties alluded to better nano-fillers dispersion and collaboration amongst the filler and the rubber. The good interfacial reaction of the rubber-GO sheets is a huge issue to withstand load by the nano-composite. These enhancements ascended because of the crosslinking improving in nano-composite structure.

The impact of GO fraction on the electrical resistivity of GO- reinforced NBR nanocomposite showed up clearly. Expansive estimations of volume resistivity acquired for most examples and no particular permeation edge was watched

even by expanding GO fraction. The present work affirmed GO/NBR nano-composite as poor conductive elastic with high resistivity values around $10^6 \Omega \text{ cm}$. The higher resistivity is likely because of the obstacle of particle development by the GO system and cross-linking change. This state decreases the charge versatility; however basically, the conductivity is relying upon the way the GO nano-sheets dispersed through the NBR.

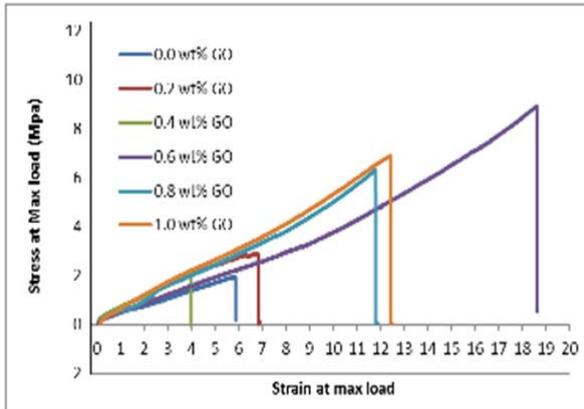


Figure 5; effects of GO existence in GO/NBR nanocomposite rubber, on Stress-strain behavior for different loading of GO.

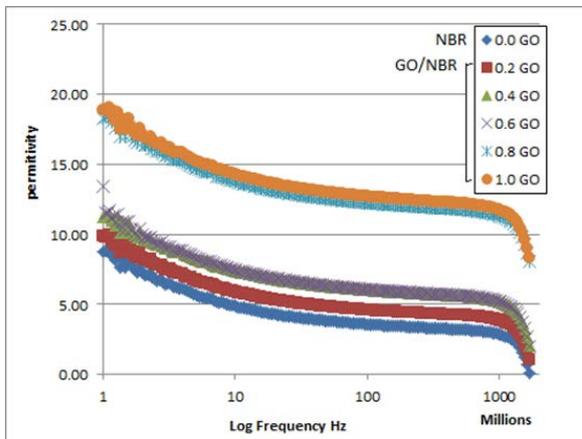


Figure 6: the dielectric property of NBR and GO/NBR between the permittivity and frequency.

In spite of the fact that GO/NBR nano-composite has low electrical conductivity, the nano-composite rubber can likewise give capacitive field evaluating from their expanded dielectric consistent. Fig. 6 demonstrates the connection amongst permittivity and Log of frequency; there is a huge change in the permittivity estimations of GO/NBR at a low frequencies contrasted and the slick NBR. However, this example diminished bit by bit at high frequencies.

Fig. 7 compares between the recent study results and several new works on the GO/NBR.

Ref. [20] investigated the properties of nitrile butadiene rubber (NBR)/polypropylene prepared by dynamical vulcanization (DV) for oil resistant. Pendinko [21] studied the effect of adding rubber carbon black or silica on the wear and mechanical properties of a hydrogenated nitrile butadiene rubber when the surface was treated with saline (SI-si and SI, respectively) and when it was used without this treatment. Zhang examined the effect of adding four types of rice bran carbon (RBC) that had different particle sizes to Nitrile-butadiene rubber (NBR) composites. Kumar studied the influence of adding four carbon based nanofillers and additional carbon black (CB) to nitrile butadiene rubber on its characteristics properties. The figure indicates reasonable results produced from our products which validates the material preparation and examinations.

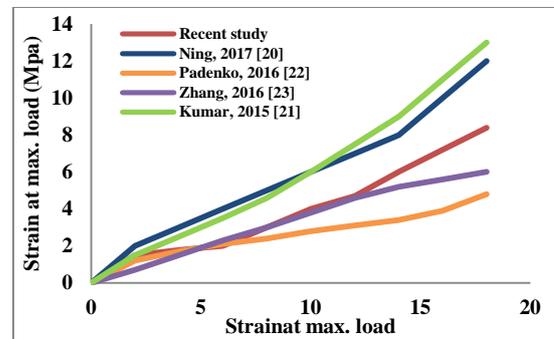


Figure 7: A comparison between the recent study results and other works on NR/NBR blends

Conclusions

The rubber networks strengthened by the nano-fillers highly affects the resulted properties. In this paper, Graphene Oxide (GO) was added and scattered in Nitrile Butadiene Rubber (NBR) and the resulted material accomplished great curing and proper attributes. The homogeneity of GO nano-sheets scattering was affirmed by the XRD Analyzing and Scanning Emission Microscopy (SEM) describing. The interacted bonds between NBR and GO brought lessened the versatility of chains, at 1.0 wt% GO fraction. The essential trademark was the presence of GO recognized by the assimilation at the same wave quantities of NBR and GO/NBR nano-composite.

The GO surface was rich in oxygen useful gatherings which made the interaction between the GO nano-sheets and NBR could be accomplished by the greatest filler rate. The rigidity of the GO/NBR nano-composite was expanded essentially when compared to NBR. The electrical conductivity of GO/NBR nano-composite indicated no obvious changes even with the ascent in the GO fraction. The dielectric property was improved partially as the permittivity has few expanding under high-

recurrence impact. The GO/NBR electrical conductivity as it is dielectric material remained low because of the improvement in the crosslinking of the nano-composite structure.

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تأثير إضافة أكسيد الكرافين في الصفات نانو-مركب مطاط بوتادين

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الخلاصة:

يمكن للدائن المقواة بواسطة الدقائق النانوية (مثل أكسيد الكرافين) ان تحسن الخواص بنسبة عالية. في هذا العمل، تم اضافة مادة اوكسيد الكرافين النانوية (GO) الى مطاط النتريل بوتادين (NBR). وتمت عملية الفلكنة للمواد النانوية المركبة (GO/NBR) باستخدام عملية المزج للحصول على انتشار متجانس لاوكسيد الكرافين في المطاط، كما ظهر واضحا عبر استخدام عملية المسح المجهرى (SEM) و اختبار الاشعة السينية (XRD). كما تم تحديد التجمعات بواسطة جهاز الأشعة تحت الحمراء (FTIR). سببت النسب الوزنية المختلفة لمادة اوكسيد الكرافين النانوية في داخل المطاط أثراً واضحاً على سلوك النانو-المركب من حيث الصلابة والمرونة. ان وجود اوكسيد الكرافين في المطاط يعمل على تطور الترابط الشبكي في التركيب الداخلي للمادة المركبة النانوية وبالتالي تؤدي الى تحسين الخواص. اضافة الى تأثيره في الموصلية الكهربائية ومرونة المادة النانوية المركبة (GO/NBR).

الكلمات الرئيسية: اوكسيد الكرافين أكسيد (GO), المطاط بوتادين النتريل (NBR), توزيع (GO) في اللدائن، مواد نانوية مركبة مطاطية (GO/NBR)