Research Article

Mechanical property evaluation of natural rubber/ vein graphite composites

M. P. Viyanage\textsuperscript{1,2}, T. R. T. Manage\textsuperscript{1,2}, R. C. L. De Silva\textsuperscript{1}, L. D. C. Nayanajith\textsuperscript{1}, M. Y. D. Milani\textsuperscript{1} and I. R. M. Kottegoda\textsuperscript{*1}

\textsuperscript{1}Material Technology Section, Industrial technology Institute, Colombo 07, Sri Lanka
\textsuperscript{2}South Eastern University of Sri Lanka, Sammanthurai, Sri Lanka.

Abstract

Vein graphite filled natural rubber composites were prepared by keeping the total weight of composites constant in each using suitable dispersant, accelerators, and coagulants to use in high-end applications. All the composites were characterized by using X-ray diffraction (XRD), scanning electron microscopy (SEM), and Fourier transform infrared spectroscopy (FTIR). The tensile properties, Young’s modulus and hardness of the composites were evaluated. XRD and FTIR analysis verified the presence of rubber and graphite in the composites without any noticeable structural changes. The SEM image of composites showed a uniform homogeneous surface of the composites. High tensile strength was observed at 10\% of graphite incorporation in natural rubber. Young’s moduli and hardness of composites were observed to be improved with the addition of the dispersed graphite. The hardness of the composites increases with increasing graphite in the composite and optimized at 60\% of graphite. The properties of natural rubber/graphite investigated in the present study is useful for many applications including electronic devices, aerospace, automobiles, toys, heavy equipment industry, battery, consumer products, etc.

Keywords: vein graphite; natural rubber; dispersion graphite

*Corresponding Author: iresha@iti.lk

DOI: http://doi.org/10.4038/sljp.v22i1.8082
1. INTRODUCTION

Recently, there is a huge demand for natural rubber (NR) worldwide in many applications. NR products are mainly classified as latex base products and dry rubber products\textsuperscript{1,2}. The common techniques for manufacturing useful products from NR latex are casting, coating, extrusion, molding, foaming, and dipping\textsuperscript{3}. Latex dipping made a wide range of rubber products. The rubber produced in the world is largely used for producing tires and related items. Pneumatic tires and tubes, solid tires, tire flaps, retread material, and puncture repair kits are the tire products. Natural rubber is the ideal base material for aircraft, heavy-duty vehicles such as buses and trucks, racing cars and tractors. NR is used in many applications because it has remarkable and desirable properties. Also, NR has superior building track, better dynamic properties, fatigue resistance, low heat build-up, better resilience, high mechanical strength, and excellent flexibility. Natural rubber excels in both abilities, making it an ideal material for dynamic seals and gaskets\textsuperscript{4}. Because of these properties, NR products are used in many fields such as engineering, sport, medical and household applications\textsuperscript{5,6}. But, for economic reasons, fillers are added into the NR to reduce the production cost by changing the volume and mass of the production. Normally, these fillers have a small effect on the mechanical and dynamic mechanical properties of the production. Also, fillers are used widely in thermoplastic and rubber industries to significantly improve modulus, tensile strength, tear resistance, abrasion resistance, and dynamic mechanical properties, etc\textsuperscript{7-9}.

Graphene, graphite oxide (GO) reveals high modulus and strength\textsuperscript{10,11}. Due to these behaviors GO is mixed with the rubber latex through the latex co-coagulation process. This composite indicated that the modulus of the NR increases with the addition of GO\textsuperscript{12}. Fillers are added in to the NR to change the properties. Normally, fillers have small effect on mechanical and dynamic mechanical properties of the products\textsuperscript{13}. Graphite is a natural raw material and has very good thermal properties. When graphite is applied as a filler to improve the properties such as thermal and electrical conductivity, the properties such as mechanical and tensile properties can be affected. All required properties for a particular application determine the quality of the final product such as solid tires\textsuperscript{14-17}.

NR in latex form with vein graphite and then NR graphite composite series have prepared. After that mechanical properties have evaluated. The homogeneous mixture of graphite was obtained through dispersion of graphite\textsuperscript{18}. 
2. EXPERIMENTAL

2.1 Materials

Natural Latex (NR), Bogala natural graphite, Acetic acid (CH$_3$COOH), Sulfur, and Stearic acid [CH$_3$(CH$_2$)$_{16}$COOH], were used as received.

2.2 Preparation of natural latex/ graphite composite dry sheets and vulcanization

Natural latex and disperse graphite were mixed for 10 minutes. After grinding well, the mixture was coagulated. Subsequently, the water is removed and dried in an oven at 105ºC to remove moisture. The composite dry sheets were prepared by changing the percentage of graphite.

Then the compounded sample was vulcanized using a hot press after adding sulfur into the dry rubber sheets.

2.3 Characterization

X-ray powder diffraction (XRD) analysis was conducted to identify the crystalline phase rubber graphite composite using Regaku ultima IV X-ray diffractometer with Cu K$_{α1}$ radiation (λ = 1.5406 Å) at 40 kV and 40 mA. The morphology of graphite, natural rubber, and natural rubber/graphite composites was characterized by LEO 1420vp scanning electron microscopy (SEM). Functional groups attached to graphite, natural rubber, and natural rubber/graphite composites were characterized using BRUKER Tenor 27, FTIR-ATR spectrometer.

The tensile properties and hardness were measured according to the ISO standards.

3. RESULTS AND DISCUSSIONS

XRD patterns of the natural graphite and dispersion graphite was depicted in figure 1. The characteristic peak for graphite was observed at 2θ=26.54° resulting from (002) reflection. Normally, graphite is composed of stack layers of carbon planes in which each plane is orderly arranged with an interlayer of carbon planes of 3.355 nm. After dispersion, the interlayer spacing of natural graphite was 3.352nm with a sharp and high peak of (002) reflecting at 2θ=26.57°. There is no significant change in graphite before and after
dispersion. Therefore, milling of graphite to prepare dispersion has not affected the crystalline nature of graphite.

![X-ray powder diffraction (XRD) patterns of vein graphite (VG) and dispersion graphite (DG).](image)

**Figure 1.** X-ray powder diffraction (XRD) patterns of vein graphite (VG) and dispersion graphite (DG).

In figure 2 the SEM images from the surface of NR/ graphite composite with 500 magnification [from figure 2 (b) - (g)] show nearly homogeneity. The milling process followed in the present study resulted in uniform mixing as a dispersant. The surfaces without imperfections were observed demonstrating the quality of particle dispersion on a polymer matrix. An SEM analysis of the prepared NR/ graphite composites was shown in figure 2. The SEM analysis of the NR (a), the NR/ graphite composites showed nearly uniform surface revealing graphite has been well mixed with NR. When (c) compared with (d), (e), (f), (g) and (h), the surface contains more graphite with increasing graphite percentage.

![SEM images](image)
The FTIR spectrums of NR and dispersion graphite were shown in figure 3(A). The FTIR spectrum of NR contains some peaks known to be characteristic of the NR structure. It showed that the absorption peaks in the NR near 3398 cm\(^{-1}\) and 1539 cm\(^{-1}\) was stretching vibration absorption peaks of the N-H group and vibration compound peak of C-N group and N-H group in the amide group, respectively. These groups were caused by proteins in NR. The characteristic bands of the saturated aliphatic \(sp^3\) C-H bonds are seen at from 2960 cm\(^{-1}\) to 2849 cm\(^{-1}\) assigned to \(\nu_{as}(CH_3)\), \(\nu_{as}(CH_2)\), and \(\nu_s(CH_2)\) respectively. The

Figure 2. SEM images of (a) NR, (b) NR/ graphite composite (10%), (c) (20%) composite, (d) (30%) composite, (e) (40%) composite, (f) NR/ graphite (50%) composite, (g) (60%) composite.
cis-1, 4-polyisoprene absorption band of the strong amplitude corresponding to =CH out of plane bending is observed at 1375 cm$^{-1}$ and it is characteristic of $CH_2$ deformation. The characteristic peak of the $-CH_3$ bending is seen at 1448 cm$^{-1}$ and $R_2C = CHR$ bonds are seen at 834 cm$^{-1}$. FTIR spectra of the composite revealed a spectrum similar to natural graphite when more graphite (50-60%) is added to rubber.

![FTIR image](image)

**Figure 3.** FTIR image of (A), (a) NR and (b) dispersion graphite, (B) NR/ dispersion graphite composite vary with graphite percentage (a) 10% graphite, (b) 20% graphite, (c) 30% graphite, (d) 40% graphite, (e) 50% graphite, (f) 60% graphite and (C), (a) dispersion graphite, (b) NR, (c) NR/ 40% graphite composite.

Figure 3 (B) shows the FTIR spectrum of NR/ dispersed graphite composite with dispersed graphite percentage. Due to similarities in the appearance of all composite, 40% composite with intensity peaks was only shown in figure 3(C). NR and dispersed graphite composite shows intensity of characteristic bands of NR chain is observed at from 2960 cm$^{-1}$ to 2850 cm$^{-1}$. Intensity of this peak was decreased with increasing graphite percentage.
Figure 4 shows the variation of the strength of the NR/graphite composite with graphite percentage. The NR/10% of graphite composite exhibits high tensile strength compared to other samples. When the graphite percentage of the composites is increased above 10% of graphite, the strength of the composites rapidly decreases. The reason is that the deformation degree of the NR rubber molecular chains under loading is restricted by the graphite. The tensile strength is related to the energy required to deform and fractures the polymer chains. The effective crosslink density of the composites increases with increase filler loading due to the polymer-filler interaction with the addition of filler. As a result, strength increases with the percentage of graphite. As a result, strength of composites that contain CNR increases up to 60% of graphite. But the strength of NR/graphite composites decreases after 10% of graphite due to the less pure latex contain.

The Yong’s moduli of the NR/graphite composites, with graphite percentage, are shown in figure 5. Comparing Young’s modulus of the composite, at the break with different percentages of graphite, it is obvious that the composites with a higher percentage of graphite show higher enhancements in the modulus. The highest modulus was observed for the composite containing 60% of graphite and 40% of NR, which was 2347.7723 Pa. The increase in Young’s modulus is due to strong interactions between the polymer chain and the graphite particle.
Figure 5. Variation of the Young Modulus of the NR/graphite composites with graphite percentage.

Figure 6. Variation of hardness of NR/graphite composite with graphite percentage.
The hardness of the NR/graphite composites at the break with graphite percentage is shown in figure 6. The composites were characteristic at room temperature. When the graphite percentage in the composite was increased, the hardness of the composite increases and a sharp increase was observed at 60% of graphite in rubber. The addition of more graphite leads to the fragile composite sheet.

![Figure 7](image)

**Figure 7.** Reinforcement mechanism of NR/Graphite composite with graphite percentage.

Figure 7 shows a natural graphite/polymer network at the low and high percentage of graphite in rubber before and after being stretched indicating indicates the mechanism for improved hardness and reduced tensile strength. The tensile strength increased with the addition of graphite up to 10% of graphite and it decreased with the further addition of graphite whereas hardness increased with increasing graphite. If low graphite amount is added the effect for tensile strength is less and at a higher amount of graphite, there are isolated graphite aggregates that not bond with rubber in the polymer network which decreases the tensile strength as well as mechanical strength. If graphite is added more than 80% then without having surrounded rubber, the composite shows fragile nature resulting in low tensile strength as well as hardness.

4. CONCLUSION

The mechanical properties of natural graphite/natural rubber composite were investigated. The graphite can be well dispersed in natural rubber by using a grinding process without any wastage. The natural rubber/graphite composites were successfully prepared with the
addition of graphite up to 60%. The highest tensile strength of the composites was found at 10% of graphite in natural rubber (NR). The Young’s moduli of the composites increased with the increasing graphite percentage. The hardness of the natural rubber/graphite composite was also increased with increasing percentage of graphite. The present investigation of mechanical properties of natural rubber/graphite composite will be very useful in applications where the combination of properties expected from graphite and natural rubber is anticipated.

ACKNOWLEDGEMENT

The work was financially supported by Treasury Grants (TG)

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