Effect of Graphite Oxide Loading on Tensile Properties of Natural Rubber Composites

W. D. M. Sampath[1,2*], C.A.N. Fernando[1], D.G. Edirisinghe [2]

Department of Nano Science Technology, Faculty of Technology, Wayamba University of Sri Lanka, Kuliyapitiya 60200 Sri Lanka[1]

Rubber Technology and Development Department, Rubber Research Institute of Sri Lanka, Telawala Road, Ratmalana 10390, Sri Lanka[2]

[*] wdmsampath@wyb.ac.lk

Abstract - Graphite-based fillers have garnered global attention in the industry and academia due to their outstanding mechanical properties such as mechanical stiffness, strength, and elasticity. In this study, a series of natural rubber (NR) composites were prepared by varying the graphite oxide (Gro) loading from 0 to 10 phr (parts per hundred rubber) at 2 phr intervals. Gro showed a significant enhancement in tensile properties and hence reinforcement of the NR composites as evident from the comparison of the results with those of the composite prepared without Gro (control). The composite prepared with 10 phr loading of Gro indicated high mechanical performance in terms of tensile strength and elongation at break. Therefore, this composite could be used in high elastic rubber-based applications. In addition, this composite shows an enhancement in tensile performance and hence, it could be economically advantageous for low volume NR based applications. Moreover, the stress-strain performance of 10 phr Gro loaded composite showed greater improvement due to representation of the largest area under the stress-strain curve and could be related to enhancement of elastic energy of the composite. Finally, the composite prepared with 10 phr loading of Gro showed about 10% increase in tensile strength compared to the control. Also, a 22% increase in elongation at break was observed in the former composite in comparison to the latter composite. Hence, the NR composite prepared with 10 phr loading of Gro could be suitable for high elastic polymeric applications requiring good modulus and tensile strength.

Keywords: Natural rubber, graphite oxide, tensile strength

I. INTRODUCTION

Graphite oxide (Gro) is generally the intermediate compound for obtaining separated graphene sheets. Further, graphite lattice has been exfoliated via chemical oxidation to disrupt the weak van der Waals forces, allowing for easy penetration with molecules. This process results in the formation of Gro, of which the interlayer space is occupied by epoxy, hydroxyl, and carbonyl functionalities [1]. Hence, Gro represents better functionality than natural graphite and this would be advantageous in improving compatibility with polymeric materials. In addition, Gro is widely used in different fields such as modern chemistry, physics, materials science, engineering, etc. [1]. Natural rubber (NR) possesses good mechanical and chemical properties, which enable NR as one of the most important raw materials used widely in the tire industry and other product manufacturing industries. NR is usually reinforced with mineral fillers in order to get substantial improvements in strength and stiffness [2]. However, the traditional reinforcing materials are not so effective in NR composites as high loadings should be incorporated to get the required properties. The most widely used filler in the rubber industry is carbon black. The incorporation of graphite-based materials such as Gro, graphene, surface-modified graphite has a potential to improve the overall performance of NR and lower loadings could be sufficient to enhance the performance of the end composite [3]. Hence Gro was used in this study, and it interacts well with the rubber when they are mixed together. In order to achieve an enhancement in properties of NR composites, fillers need to be well dispersed and homogenized with the rubber.

II. MATERIALS AND METHODS

A. Materials

RSS-2 (Ribber Smoked Sheet rubber) was supplied by the Rubber Research Institute of Sri Lanka. Graphite having a mean particle size of 14 micron was obtained from Bogala Graphite Lanka PLC., Sri Lanka. All rubber compounding ingredients were purchased from local suppliers. Potassium permanganate ($KMnO_4$), ethanol (C_2H_5OH), hydrochloric acid (HCl), sulfuric acid (H_2SO_4), and hydrogen peroxide (H_2O_2) were purchased from Organic Trading (Pvt.) Ltd., Sri Lanka.

B. Methodology

Preparation of NR composites filled with Gro

A series of NR composites was formulated by varying the Gro loading from 2 to 10 phr at 2 phr intervals. The NR composite prepared without Gro was considered as the control. The formulation of the composites is given in **Table 1**. The composites were prepared by melt mixing using a Brabender Plasticorder operated at room temperature, at a rotor speed of 60 rpm. Total mixing time was kept constant at 10 min.

Table 1. Formulation of NR composites filled with Gro

Ingredient	Function	Phr
NR	Rubber	100
ZnO	Inorganic activator	5.0
Stearic acid	Organic activator	2.0
TMQ	Antioxidant	1.0
Gro	Filler	0 2 4 6 8 10
ZDC	Accelerator	1.5
Sulphur	Vulcanizing agent	2.0

Determination of tensile properties

Tensile properties of Gro/NR composites were determined using Instron tensile testing machine according to BS ISO 37:2017. Dumb-bell shaped tensile test specimens were used. Cross-head speed was maintained at 500 mm/min with three replicates per treatment.

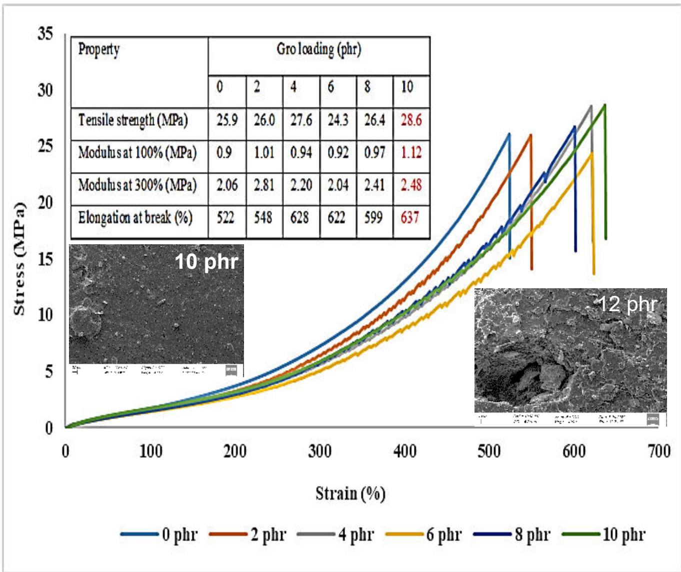
III. RESULTS AND DISCUSSION

Stress-strain curves provide an extremely important graphical measure of mechanical properties of a material such as modulus, tensile strength and elongation at break. These parameters are highly important to explain the elastic behavior of a polymeric material and special rubber-based materials. **Fig. 1** shows the stress-strain behaviour of NR composites prepared with and without Gro. The composite prepared with

10 phr loading of Gro shows higher stress-strain properties compared to the other Gro filled composites and the control. Generally, toughness of rubber composites increases with filler loading [4]. The 10 phr Gro loaded composite shows the highest toughness, which is indicated by the highest area under the stress-strain curve. In addition, area under this curve represents the elastic potential energy of polymeric materials. Hence, the composite prepared with 10 phr loading of Gro shows a higher elastic potential energy than the other composites. Tensile strength of most of the Gro loaded composites is higher than that of the control. The composite prepared with 10 phr loading of Gro shows the highest tensile strength (28.6 MPa) and it could be attributed to greater toughness. In contrast, the composite prepared with 6 phr loading of Gro shows the lowest tensile strength (24.3 MPa) and the reason might be due to agglomeration of graphite-based materials at lower loading levels [5]. All the composites exhibit similar tangents at 100% elongation and hence, modulus at 100% elongation of NR composites has not shown a significant variation with the increase of Gro loading (Figure 1). Further, modulus at 100% elongation indicates hardness of a rubber material. Hence, the composite prepared with 10 phr loading of Gro indicates the highest hardness out of all the composites. However, all the Gro-filled NR composites show a higher modulus at 100% elongation compared to the control. The factors that affect reinforcing potential of fillers include filler dispersion, surface reactivity, bonding strength between the rubber and the filler, etc. Variation of modulus at 300% elongation of NR composites with Gro loading is also shown in Figure 1. Modulus at 300% elongation of all six composites is observed in the range 2.04 – 2.48 MPa. Modulus at 300% elongation of most of the composites prepared with Gro is higher than that of the control. The composite prepared with 10 phr loading of Gro shows the highest modulus at 300% elongation and it can be attributed to the highest crosslinking density. Elongation at break of all composites is higher than 500% (Figure 1) and it is a good indication of elastic performance. Elongation at break of all Gro filled NR composites is higher than that of the control and this can be attributed to better dispersion of Gro in the NR matrix and further, better reactivity of Gro with NR via the hydroxyl group. The composite prepared with 10 phr loading of Gro shows the highest elastic properties, and the reason may be better adhesion between the NR phase and the functionalized Gro. Moreover, the composite prepared with 12 phr loading of Gro illustrates poor surface morphology (Fig. 1) compared to 10 phr loading Gro. Hence, 10 phr loading of Gro would be used as optimum loading to enhance the tensile properties of NR composites.

IV. Conclusions

The composite prepared with 10 phr loading of Gro showed 10.4 % increase in tensile strength compared to the control. Further, the Gro filled NR composites showed a remarkable improvement in elongation at break in comparison to the control. Hence, it can be concluded that the NR composite prepared with 10 phr loading of Gro could be suitable for high elastic polymeric applications requiring good modulus and tensile strength.



Property	Gro loading (phr)					
	0	2	4	6	8	10
Tensile strength (MPa)	25.9	26.0	27.6	24.3	26.4	28.6
Modulus at 100% (MPa)	0.9	1.01	0.94	0.92	0.97	1.12
Modulus at 300% (MPa)	2.06	2.81	2.20	2.04	2.41	2.48
Elongation at break (%)	522	548	628	622	599	637

Fig. 1 SEQ Figure * ARABIC 2. Variation of stress-strain properties of NR composites with Gro loading

References

[1] C.P. Tien and H. Teng, “Polymer/graphite oxide composites as high-performance materials for electric double layer capacitors”. *Journal of Power Sources*, *195*(8), pp.2414-2418, 2010.

[2] L.O. Ekebafe , Adebayo, M.N. Chukwu, K.O. and Eguare “Performance evaluation of graphite oxide in natural rubber compounds”. *Nigerian Journal of Chemical Research*, *24*(1), pp.20-33, 2019.

[3] W.D.M. Sampath, C.A.N. Fernando and D.G. Edirisinghe, “Synthesis and characterization of poly (ethylene glycol)-grafted graphite and graphite oxide and comparison with natural graphite”. In 10th YSF Symposium (p. 194), March 2022.

[4] L. Bokobza, “Mechanical and electrical properties of elastomer nanocomposites based on different carbon nanomaterials”. *C—Journal of Carbon Research*, 3(2), p.10, 2017.

[5] W. Kitisavetjit, Y. Nakaramontri, S. Pichaiyut, S. Wisunthorn, C. Nakason and S. Kiatkamjornwong, “Influences of carbon nanotubes and graphite hybrid filler on properties of natural rubber nanocomposites” *Polymer Testing*, 93, p.106981, 2021.