

EXPERIMENTAL STUDY ON MECHANICAL PROPERTIES OF ELASTOMER CONTAINING CARBON NANOTUBES

R. ISMAIL^{1,2,*}, A. IBRAHIM², H. HAMID², M. RUSOP³, A. ADNAN⁴

¹Institute for Infrastructure Engineering and Sustainable Management (IIESM),
Universiti Teknologi MARA (UiTM), 40450, Shah Alam, Selangor, Malaysia

²Faculty of Civil Engineering, Universiti Teknologi MARA (UiTM),
40450, Shah Alam, Selangor, Malaysia

³NANO-ElecTronic Centre (NET), Faculty of Electrical Engineering, Universiti Teknologi
MARA (UiTM), 40450, Shah Alam, Selangor, Malaysia

⁴Faculty of Civil Engineering, Universiti Teknologi Malaysia (UTM),
81310, Skudai, Johor, Malaysia

*Corresponding Author: rozaina_fka_uitm@yahoo.com

Abstract

Recently, elastomer reinforced with nanofillers have attracted great interest due to their properties. The incorporation of carbon nanotubes into elastomers improves significantly their mechanical and dynamic mechanical properties. Carbon nanotubes (CNTs) were used to prepare natural rubber (NR) nanocomposites. Four different NR compounds containing CNTs were investigated. Compounds were prepared by a two roll mill with conventional and efficient cure systems. The properties of the nanocomposites such as tensile strength, tensile modulus and elongation at break were studied. Results obtained show that a smaller amount of CNTs can effectively improve the performance of NR. NR with 1% CNTs composites exhibited better tensile strength compared to other compounds. The study also indicated that filler materials effect on the mechanical properties of the blends.

Keywords: Tensile properties, Carbon nanotubes (CNTs), Natural rubber,
Nanocomposite,

1. Introduction

The effect of earthquakes has been one of the major concerns of scientists and engineers for a long time. Many studies were conducted on mitigating seismic responses of structures due to earthquake events [1-4]. Elastomers are invariably reinforced with nanofillers to improve their physical and mechanical

properties of the final product [5]. The mechanical and physical properties of NR were very excellent such as tensile strength, tensile modulus, tear strength and hardness [5-7]. The use of fillers nowadays are focusing on to modify and the mechanical, electrical and/or optical properties of rubber compounds[8]. However, NR is sensitive material when exposed to solvents, deteriorated by oxygen, ozone, sun light, UV rays as well as humidity [9,10]. The effect of fillers to the rubber compounds has been performed by several authors [6, 7] to study the mechanical and dynamic mechanical properties. The chemical and physical properties of nanocomposites were greatly restricted and were significantly enhanced [11-16].

2. Experimental

2.1. Materials

Table 1 presents the materials that were used in compounding NR nanocomposites. Natural rubber SMR 10 was supplied by Lembaga Getah Malaysia (LGM). Carbon black filler type SAF N10 was used. Other material such as zinc oxide, stearic acid and sulphur are required as the basic ingredients of compounding unfilled rubber or filled rubber. Antioxidant, N-cyclohexyl-2-benzothiazole sulphenamide (CBS) and tetramethylthiuram disulphide (TMTD) are additives that have been selected in order to increase the properties of rubber. Moreover, each of them has specific function.

Table 1. Materials in NR nanocomposites.

Materials	Description
Natural Rubber	Type: SMR 10
Carbon Black	SAF N110
Carbon Nanotubes	Diameter <50nm, Length~20 μ m, purity>95wt%
Activator	Zinc oxide
Co-activator	Stearic acid
Additives	Antioxidant
Vulcanizing agent	Sulphur
CBS	N-Cyclohexyl-2-benzothiazole sulfonamide
TMTD	Tetramethylthiuram Disulphide

2.2. Compounding and cured characteristics

Mixing of rubber compound was performed according to ASTM D 3182 [17] by using a laboratory open two-roll mill (400 mm diameter and 600 mm working length). The mixing process needs to be conducted carefully and the right sequence has to be followed in order to get the finest elastomer compounding. It is essential to be assured that the molecular of the crude rubber is break before adding in other ingredients. The rubber mixes obtained was in sheet left for a period at least 16 hours before testing. All the ingredients were added and carried out by following the same manner and conditions of mixing. All compounds had the same composition, except for the amounts of CNTs (0%, 1%, 3% and 5%). The compounding formulations (in parts per hundred parts of rubber (phr)) used in this work are listed in Table 2.

Table 2. The formulation for rubber compounds.

Ingredients (pphr)	0 CNTs/ NR	1 CNTs/ NR	3 CNTs/ NR	5 CNTs/ NR
Natural Rubber (SMR 10)			100	
Carbon Nanotubes (%)	0	1	3	5
Carbon Black (SAF) N110			25	
Antioxidant			2	
Zinc Oxide (ZnO)			5	
Stearic acid (St.Acid)			2	
TMQ			2	
Sulphur			1.5	
CBS			0.5	
TMTD			0.5	

2.3. Vulcanization

In general, the manufacturing of rubber products includes compounding, mixing, processing, assembly and vulcanization. The main processing operations involve extrusion for receiving shape, or calendaring to sheet material. Vulcanization converts the essentially plastic rubber mixture to an elastic state. Vulcanization is accomplished by heating the shaped rubber. The required heat is usually transferred by the metal of the mold in which the rubber is shaped. The compounds were compression-moulded using a hot press machine at 150°C to the respective cure times of compounds [18].

2.4. Testing

Tensile test provides the tensile stress-strain properties include tensile strength, tensile modulus, and elongation at break [19]. Tensile properties of specimens were measured according ASTM D 412 [20], dumbbell shaped samples were cut from the mould sheets. Tear properties of specimens were determined according ASTM D 624 [21] with trousers test pieces. The crosshead rate for tear test was 100 mm/min. The dumbbell test pieces were tested by using an Instron Universal Tensile Machine equipped with 500N load cell at a static crosshead speed of 500 mm/min according to BS ISO 37:2011 [22]. All specimens have been tested at a stretching rate of 500 mm/min by using a Tensile Instron Machine. The indentions of hardness of these four compounds were measured by a Wallace Dead Load Tester according to the method described by BS ISO 48:2010 [23]. A Wallace Dunlop Tripsometer was used to determine the rebound of test piece by referring to BS 903-A8:1990 [24]. Fig. 1 shows a dumb-bell test-piece. Details of test-piece preparation and test conditions are described in BS ISO 37:2011 [22].



Fig. 1. Dumbbell test piece.

3. Results and discussion

The tensile strength, the stress at 100% and 300% strain; M100 and M300, elongation at break, hardness and rebound were then determined by standard methods. The result obtained show that the physical properties of NR are affected by the amount of CNTs incorporated into the rubber and the way the nanocarbon is incorporated into the rubber composition.

3.1. Tensile strength

Tensile modulus as applied to the elastomers is defined as the force require to produces a certain elongation. Tensile strength was measured by using a tensile machine in accordance with ISO 37. Table 3 shows that rubber compounds of that five different percentage of CNTs are strongly increased modulus at 100 and 300% elongation from 0pphr up to 3pphr, however the percentages was a bit lower for 5% CNTs. In addition tensile modulus measure the stiffness and vulcanization degree of rubber compounds. The tensile modulus at 100%, and 300% was steadily increased. It can be seen that CNTs with 3pphr with the amount 1.66 MPa and 6.37 MPa was the highest tensile modulus at 100% and 300% respectively.

Table 3. The Properties of NR containing different percentage of CNTs.

Physical Properties	Amount of CNTs (pphr)			
	0	1	3	5
Tensile Strength (MPa)	27.09	29.32	28.84	27.90
Modulus M100 (MPa)	1.28	1.42	1.99	1.27
Modulus M300 (MPa)	5.37	5.54	6.37	3.11
Elongation at Break (%)	818.28	974.16	816.61	312.46

Figure 2 shows the composition with 1pphr of nanocarbon gave greater tensile strength and the higher elongation at break than the comparative compositions with 0, 3 and 5 pphr of nanocarbon.

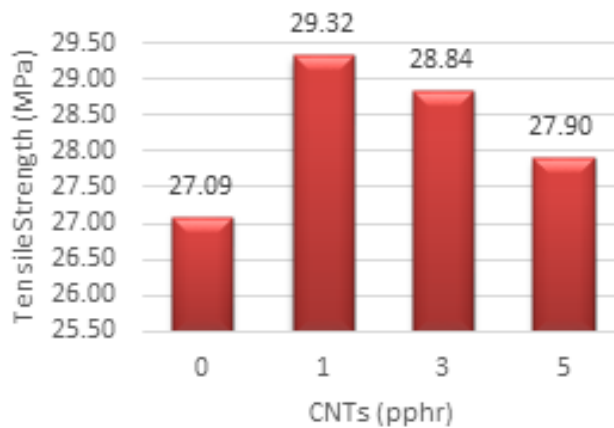


Fig. 2. Effect of different amount of CNTs on tensile strength.

The elongation at break means the elongation at the time of rupture of a rubber compound. Form Fig. 3, the best elongation at break among samples was 1pphr

with the value of 974.16%. NR without CNTs experienced 818.28 % elongation. However, the decreasing trend shows start from 1pphr followed by 2pphr and 3pphr with values of 816.61% and 312.46% respectively.

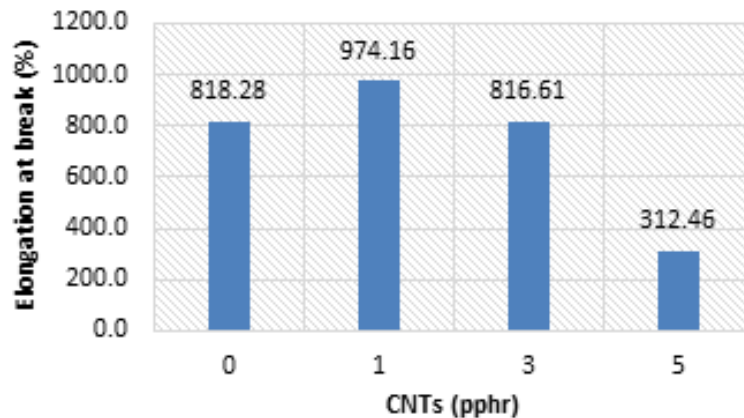


Fig. 3. Effect of different amount of CNTs on elongation at break.

The incorporation of carbon nanotubes into elastomers improves significantly when the contents of carbon black from the conventional compound was replaced the nanocarbon. Table 4 shows the excellent performance of CNT/NR nanocomposite with high tensile strength compared to the conventional carbon black/ NR. Tensile strength of sample containing CNTs is 81.27 MPa shows that 383% higher than 16.82 MPa with the present of carbon black.

Table 4. Tensile strength of NR nanocomposite.

	Tensile Strength (MPa)
CNT/NR	81.27
Carbon Black/NR	16.82

3.2. Hardness

The International Rubber Hardness Degrees (IRHD) test method is originated in Europe. It provides a very repeatable result on rubber parts of various shapes and sizes. Referring to the graph, the increasing pattern of hardness results can be seen in Fig. 4. The lowest value of hardness starts from 0% of nanocarbon with a value of 54.0 IRHD, followed by 1%, 3% and 5% nanocarbon with values of 57.5 IRHD, 61.0 IRHD and 62.5 IRHD respectively. Thus, the addition of nanocarbon gives a gradual increment on hardness of rubber. As more filler added leads to more rigid elastomer composites. Also, results in higher modulus elastomers.

3.3. Rebound resilience

Resilience of a rubber compound is a measurement of how elastic it is when exposed to various stresses. Resilience is the ratio of energy released in deformation recovery to the energy that caused the deformation. The rebound values of four rubber compounds with varies addition of nanocarbon were

compared as in Fig. 5. The addition of nanocarbon with the amount of 1% pphr increases. The addition of CNTs from 1% pphr to 5% pphr lead to decreasing values of rebound. These results explained the viscoelastic behaviour of elastomer, where energy loss is due to internal friction in the rubber itself. Less rebound value shows low dissipation of energy during recovery. This phenomenon is significant to be implemented in elastomer, which requires low damping properties.

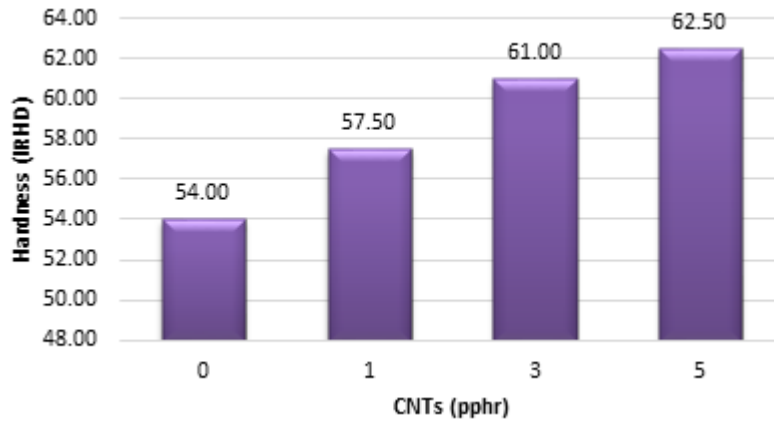


Fig. 4. Effect of different amount of CNTs on hardness.

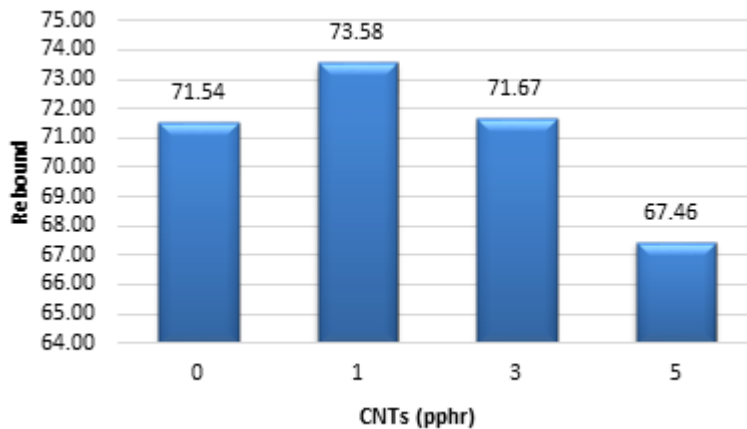


Fig. 5. Effect of different amount of CNTs on rebound.

3.4. Morphology study

Fig. 6 shows field emission scanning electron microscopy (FESEM) micrograph of CNTs) which reveals that the CNTs particles are well dispersed, thus contributing to a greater tensile strength, tear strength and hardness of the CNTs filled rubber are taken for study. Even though the interparticles of CNTs and carbon black are not clear, the addition of CNTs has significantly give effect to the microstructure of elastomer.

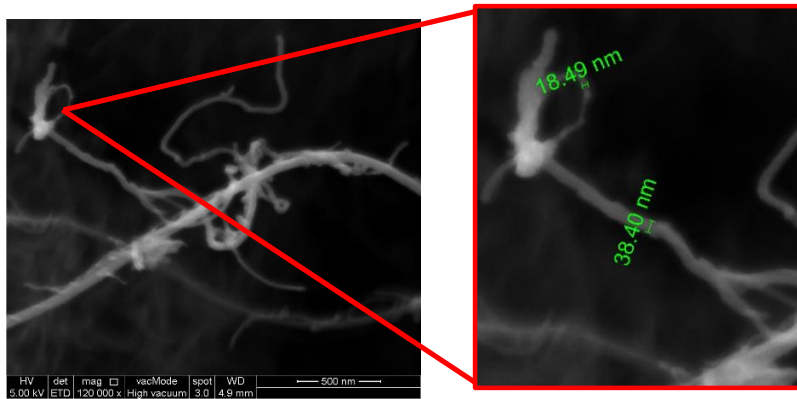


Fig. 6. FESEM micrograph of CNTs at 120000 magnification.

4. Conclusions

In conclusion, the properties of nanocomposites depend greatly on the chemistry of polymer matrices, nature of nanofillers, and the method in which they are prepared. The incorporation of carbon nanotubes into elastomers improves significantly their mechanical and dynamic mechanical properties. This study was show that the different filler and loading give the different effect reinforcing to rubber compound. The results obtained show that a smaller amount of CNTs can effectively improve the performance of NR. NR with 1% CNTs composites exhibited better tensile strength compared to other compounds. The study also indicated that filler materials effect on the mechanical properties of the blends and show that the significantly improved performance of nanocomposites.

References

1. Ismail, R.; and Ismail, M.I. (2017). Dynamic analysis of concrete faced rockfill dam using finite element method. *Journal of Engineering and Applied Sciences*, 12(7), 1772-1776.
2. Ismail, R.; and Abdul Karim, M.R. (2017). Concrete bridge pier performance under earthquake loading. *Journal of Engineering and Applied Sciences*, 12(9), 2254-2258.
3. Ismail R.; Ibrahim, A.; and Razali, N. (2017). Vulnerability study of public buildings subjected to earthquake event. *MATEC Web Conference*, 103, 02023.
4. Ismail, R.; Kamsani, M.H.; and Mohd Nadzri, N.I. (2017). Seismic analysis of concrete dam by using finite element Method. *MATEC Web Conference*, 103, 02024.
5. Blow, C.M.; and Hepburn, C. (1982). Rubber technology and manufacture, *The Plastics and Rubber Institute*.
6. Sangwichien, C.; Sumanatrakool, P.; and Patarapaiboolchai, O. (2008). Effect of filler loading on curing characteristics and mechanical properties of thermoplastic vulcanizate. *Chiang Mai Journal of Science*. 35.

7. Rajhan, N.H.; Ismail, R.; Abdul Hamid, H.; and Ibrahim, A. (2014). Influence of different fillers on the tensile properties of 50/50 NR/NBR blend. *Proceeding of Proceedings of the International Civil and Infrastructure Engineering Conference*, Springer Singapore, 767-777.
8. Mujkanovi, A.; Vasiljevi, L.; and Ostoji, G. (2009). Non-black fillers for elastomers. *13th International Research/Expert Conference Trends in the Development of Machinery and Associated Technology*, 865-868.
9. Vinod, V.S.; Varghese, S.; and Kuriakose, B. (2002). Degradation behaviour of natural rubber aluminium powder composites: effect of heat, ozone and high energy radiation. *Polymer Degradation and Stability*, 75, 405-412.
10. Indian Rubber Institute (1998). *Rubber Engineering*, Tata McGraw-Hill.
11. Ismail, R.; Ibrahim, A.; Abdul Hamid, H.; Mahmood, M.R.; and Adnan, A. (2014). Performance of carbon nanotubes (CNT) based natural rubber composites: A review. *Proceedings of the International Civil and Infrastructure Engineering Conference*, Springer Singapore, 821-829.
12. Chen, Y.; Peng, Z.; Kong, L.X.; Huang, M.F.; and Li, P.W. (2008). Natural rubber nanocomposite reinforced with nano silica. *Polymer Engineering and Science*, 48(9), 1674-1677.
13. Mokhtari Motameni, M.; Kokabi, M.; and Bahramian, A.R. (2013). The effect of shear strain amplitude and loading cycle on the horizontal characteristics of fiber reinforced nanocomposite elastomeric seismic isolators. *International Polymer Processing*, 28(2), 168-173.
14. Li, Y.; Li, M.; Pang, M.; Feng, S.; Zhang, J.; Zhang, C. (2015). Effects of multi-walled carbon nanotube structures on the electrical and mechanical properties of silicone rubber filled with multi-walled carbon nanotubes. *Journal of Materials Chemistry C*, 3(12), 5573-5579.
15. Jeyaraj Pandian, J.; and Govindan, T. (2015). Comparative studies on the mechanical properties of natural rubber and natural rubber carbon black composites. *International Journal of Emerging Technology in Computer Science & Electronics (IJETCSE)*, 13(2), 48-52.
16. Boonbumrung, A.; Sae-oui, P.; and Sirisinha, C. (2016). Reinforcement of multiwalled carbon nanotube in nitrile rubber: in comparison with carbon black, conductive carbon black, and precipitated silica. *Journal of Nanomaterials*, 2016, 1-8.
17. ASTM D3182-16. (2016). Standard practice for rubber-materials, equipment, and procedures for mixing standard compounds and preparing standard vulcanized sheets. *ASTM International*, West Conshohocken, PA.
18. BS ISO 3417. (2008). Rubber-measurement of vulcanization characteristics with oscillation disk curemeter. *British Standards Publication*.
19. Samsuri, A. (2009). An introduction to polymer science and rubber technology. *Pusat Penerbitan Universiti*, Universiti Teknologi MARA, 2009, 270.
20. ASTM D412-16 (2016). Standard test methods for vulcanized rubber and thermoplastic elastomers-tension. *ASTM International*, West Conshohocken, PA.
21. ASTM D624-00 (2012). Standard test method for tear strength of conventional vulcanized rubber and thermoplastic elastomers. *ASTM International*, West Conshohocken, PA.

22. BS ISO 37 (2011). Rubber, vulcanized or thermoplastic-Determination of tensile stress-strain properties. British Standards Publication.
23. BS ISO 48 (2010). Rubber, vulcanized or thermoplastic - Determination of hardness (hardness between 10 IRHD and 100 IRHD). British Standards Publication.
24. BS 903-A8 (1990). Physical testing of rubber-Part A8: Method for determination of rebound resilience. British Standards Publication.