ASEAN Engineering Journal

Full Paper

REINFORCEMENT OF CHARCOAL ACTIVATED CARBON (CAC) IN NATURAL RUBBER (NR) COMPOUND: IN COMPARISON WITH CARBON BLACK

Sharifah Nafisah Syed Ismail[a]*, Nik Noor Idayu Nik Ibrahim[b], Siti Nabila Rasli[a], Noor Aishatun Majid[a], Nor Mazlina Abdul Wahab[a], Siti Noorashikin Jamal[a], Salamiah Zakaria[a], Khuzaimah Nazir[a]

[a]Fakulti Sains Gunaan, Universiti Teknologi MARA, Cawangan Perlis, Kampus Arau, 02600, Arau, Perlis, Malaysia, Kuala Lumpur, Malaysia
[b]Faculty of Applied Sciences, Universiti Teknologi MARA, 40450 Shah Alam, Selangor, Malaysia

Article history
Received
24 June 2021
Received in revised form
13 September 2021
Accepted
01 December 2021
Published online
31 May 2022

*Corresponding author
sharifahnafisah@uitm.edu.my

Graphical abstract

Charcoal activated carbon

Abstract

The physical and mechanical properties of natural rubber (NR) filled with Charcoal Activated Carbon (CAC) and Carbon Black (CB) were studied. NR is a high-quality, bio-based material that has a more environmentally friendly production system than synthetic rubbers. The incorporation of reinforcing fillers into the natural rubber can significantly improve the mechanical properties, and up to now, CB has been one of the widely used filler. CAC had its own set of benefits for polymer engineering applications, particularly as a potential new natural-based filler in rubber composites. SMR-10 was compounded with compounding ingredients using a two-roll mill by Conventional Vulcanization system. As a comparison study, commercial grade CB (N220) filled NR was compounded alongside with the CAC/NR at 10 phr and 15 phr (parts per hundred of rubber) loading. A cure test was performed to determine the scorch time (t_{s2}) and the cure time (t_{c90}). The density, tensile strength, M100 and M300 of CAC/NR were lower than CB/NR due to the poor interaction of CAC-rubber and the possibility of slightly polar. However, the mentioned properties showed a promising increment as the filler loading increased. The swelling index (%) of CAC/NR were higher which might be contributed by the porous structure of CAC that assisting in the penetration of the toluene. As a conclusion, CAC has the potential to be used as reinforcing filler for elastomer due to the porous structure which can provide greater surface area for interaction. However, if compared to CB, higher loading of CAC was required to obtain the properties of CB of lower loading.

Keywords: Carbon black, charcoal activated carbon, compounding, natural rubber, filler

1.0 INTRODUCTION

Harvested from the Hevea Brasiliensis tree in the foam of latex, natural rubbers are one of the important industrial materials that are widely used in a variety of fields due to their high elasticity [1]. Hevea Brasiliensis is a native Brazilian plant which now cultivated mostly in Southeast Asian countries like Malaysia, Thailand, and Indonesia. Beside Hevea Brasiliensis, other plants such as rubber fig. Panama rubber, Congo rubber (Landolphia Owariensis) and the common dandelion are also sources of the natural rubber. However, Hevea Brasiliensis species is preferred due to the well growth under cultivation [2,3]. Natural rubber (NR) is a high-quality, bio-based material that has a more environmentally friendly production system than synthetic rubbers [4]. In general, natural rubber is known for its strong, flexible, and heat-resistance properties. Despite of the advantages of the natural rubber, it is still lower in term of the resistance to abrasion and ageing compared to synthetic rubber.

The incorporation of reinforcing fillers into the natural rubber can significantly improve the barrier and mechanical properties, wear and friction, and up to now, carbon black (CB) has been one of the fillers used to prepare polymer composites with high exhibitions. According to Thaptong et al. [5], using CB as a filler in rubber compounding can increase ultimate strength, modulus, and abrasion resistance, and silica being the most competitive reinforcing filler to carbon black. However, the utilization of silica as filler in non-polar rubber is limited because it is difficult to process due to the incompatibility and resulted in weak rubber-filler interaction. Free silanol groups on the surface of the silica's particle are acidic in nature and have high tendency to absorb the accelerators which led to the retardation of vulcanization process.

On the other hand, despite of proven to effectively improve mechanical performance of the tire tread [6], CB is manufactured using depleting petroleum resources, which can, in turn, contribute to the greenhouse effect [7]. According to Ahmed N. A. [8], increased percentage loading of CB in SMR20 resulted in the improvement of Shore A hardness and decrement in wear rate which indicating the enhancement in the wear resistance. The reinforcing mechanism of carbon black in natural rubber is a synergistic effect of a few factors. The magnitude of reinforcement is normally influence by the particle size, structure, specific surface area, and surface chemistry. B. Omes et al. [9] found that the Young's modulus of tension and compression for two different types of CB reinforced NR were increased as the filler volume fraction increased. However, at the same range of volume fraction, the NR reinforced with filler of smaller particle's size showed higher Young's modulus. This might be contributed by the higher specific surface area possessed by the smaller particle's size filler which provide more surface for interaction with vicinity NR leading to the higher enhancement in the mechanical properties.

While CB is produced through a complex set of reactions between hydrocarbon fuel and preheated air inside a reactor at a temperature up to 1800°C, charcoal is made from carbonised woods or animal bones which primarily used as a fuel. Charcoal is not only useful as a source of energy, but it is also having the potential to be converted into activated carbon via the activation method. According to Meng et al. [10], the recent volatility in the price of petroleum has increased the appeal of incorporating filler from renewable resources with polymeric materials rather than using reinforcing filler. As a result, bamboo charcoal powder was used to replace conventional filler in styrene butadiene rubber, with the temperature of thermal decomposition steadily increasing as the thermal compound's stability improved. By investigating its cure behaviour and mechanical properties, this study aims to compare the reinforcement efficiency of charcoal activated carbon (CAC) with conventional reinforcing filler, CB, in the sulphur vulcanization system of NR.

2.0 METHODOLOGY

Materials and Methods

Materials

CAC was supplied by Classic Chemicals Sdn. Bhd. and was used as reinforcing fillers. SMR-10 was obtained from Mardec Polymer Sdn. Bhd. Carbon black (N220), Zinc Oxide, Stearic Acid, polymerized 2,2,4-trimethyl-1,2-dihydroquinoline (TMQ) antioxidant, Mercaptobenzothiazole (MBT) accelerator and sulphur were purchased from Sigma Aldrich Sdn. Bhd.

Rubber Compounding

A two-roll mill was used for rubber compounding. SMR-10 natural rubber was masticated for five minutes using a mastication process. Following that, other compounding ingredients such as Zinc Oxide, Stearic Acid, TMQ, CB or CAC filler, MBT, and finally sulphur were added. The compounding procedure was completed in 25 minutes. For the masticating and mixing processes, the nip gap was 3 mm, 2 mm, and 0.6 mm, respectively, while the homogenize process used a 1.2 mm nip gap. The compounds were sheeted out by passing the rolled batch six times endwise through the mill nip. The vulcanization system used was a traditional system. Table 1 shows the compound formulation.

Table 1 Formulation of rubber compound

Ingredients (phr)	10 CAC	15 CAC	10 CB	15 CB
NR (SMR 10)	100	100	100	100
Zinc Oxide	5	5	5	5
Stearic Acid	2	2	2	2
TMQ	2	2	2	2
CAC	10	15	-	-
CB	-	-	10	15
MBT	1	1	1	1
Sulphur	2.5	2.5	2.5	2.5

Curing Characteristics

The curing time was investigated using a Moving Die Rheometer (Hung Ta Instrument Go. Ltd). A piece of uncured rubber was placed inside the curemeter as a test. A cure test was performed on an oscillating disc rheometer at 170°C in accordance with ASTM D-2084. The rubber samples were cut into squares weighing 6 to 8 gram and then wrapped in plastic film. The scorch time (t_{S2}), and optimum cure time (t_{C90}) are determined by rheometric analysis. The cure rubber index (CRI) was calculated using Equation (1).

$$CRI\ (1/min^{-1}) = 100 / (t_{C90} - t_{S2}) \quad (1)$$

Physical Testing

Density Measurement

The density measurement was performed in accordance with ASTM D1817. The weight of the samples for the density measurement ranged between 1 and 1.5 gram. Each sample

was weighted and recorded prior to the density test. The samples were then immersed in water, and the density readings were recorded. The density measurement was performed on five samples, and the result is an average value.

Hardness Test

Hardness test Shore A was performed on Hilderbrand hardness tester according to ASTM D2240. International Rubber Hardness Degrees are used to express the hardness value (IRHD). The reading was taken in five different locations on the specimen, and the average was computed. Within 1 second of the pressure foot making firm contact with the specimen, the durometer hardness was measured.

Swelling Test

Swelling tests were performed in accordance with ASTM D471, with the sample measuring 10 mm in length, 10 mm in width, and 2 mm in thickness. The sample was weighted prior to the swelling test, and the reading was recorded as W_{un}. The sample was then immersed in the toluene solution before being sealed in the container. The sample was removed from the solution and weighed after 24 hours. The reading was taken, and the sample was returned to the solution. These steps were repeated every 24 hours until a consistent weight reading of the sample was obtained, and the value was recorded as W_{sw}. The swelling index, Q_s, was calculated using Equation (2).

$$Q_s\ (\%) = (W_{sw} - W_{un})/\ W_{un}\ \times 100 \qquad (2)$$

Mechanical Testing

Tensile Test

Tensile testing was carried out according to ASTM D412 by using Universal Instron Tensile Machine (2.0 kN loadcell) with a crosshead speed of 500 mm/min and a gauge length of 25 mm. The samples were cut into dumbbell-shaped (2 mm thickness x 6.0 mm width) and each composition was tested with five identical samples. Tensile properties such as tensile strength, tensile modulus, and elongation at break were recorded and averaged.

3.0 RESULTS AND DISCUSSION

Cure Characteristics

Table 2 listed the scorch time (t_{S2}), optimum cure time (t_{C90}), and the CRI for the 10% and 15% CAC and CB. The scorch time indicate the working range of rubber before the curing process begin. The increment of CAC loading from 10 phr to 15 phr resulted in the increased of t_{S2} from 5.06 mins to 5.39 mins whereas the value of t_{S2} for 15 phr of CAC is similar to the value of t_{S2} of 10 phr CB. Increment of the t_{S2} from 10 phr to 15 phr of CAC might suggested the poor interaction between CAC and vicinity rubber matrix [11]. However, as the loading of CAC was decreased to 15 phr, the t_{C90} and CRI increased. t_{C90} is the curing time required for the rubber composite to fully cure. As shown in Table 2, the cure time for CB filler loading was also on the decline. The filled vulcanizates' cure time was reduced because the rubber compounds spent more time in the mill during mixing. As the filler loading increases, the incorporation time increases, resulting in more heat generated due to friction [12]. As a result, as the amount of CAC and CB increases, the time taken for compounding process increases, thus, heat and energy produced are also increased.

The reduction of t_{C90} for both CAC and CB at 15 phr indicates that there is an increment in the development of heat and energy. According to Ahmed [13], during compounding, high energy and heat are produced, causing the viscosity of the rubber compound to increase with better shear heating, resulting in a decrease in the t_{C90} compounds.

Table 2 Scorch Time, Cure Time and Cure Rubber Index of CAC/NR and CB/NR

Filler loadings	t_{S2}	tC_{90}	CRI (1/min^{-1})
10 CAC	5.06	7.61	39.20
15 CAC	5.39	7.30	52.40
10 CB	5.40	5.89	204.10
15 CB	5.09	5.79	142.90

Physical Properties

Density

Figure 1 shows the result of density for 10 phr and 15 phr of CAC and CB. 15 CB has the highest density of 1.478 g/cm^3. At both filler loading, the density of CB/NR were higher compared to the CAC/NR. This might be contributed by the porous structure of the CAC itself and the poor filler-rubber interaction as suggested by the t_{S2}. According to Zhu et al. [14], the interface bonding strength between the particles and matrix influences the enhancement of a granular filling system with bamboo charcoal (BC) hexagonal molecular structure and porous structure. Furthermore, CB is a non-polar filler which make it more compatible with the hydrocarbon rubber chain, resulting in good contact with the rubber and making the rubber composite less porous. A greater rubber-filler interaction is required to maximize filler dispersion and provide reinforcement, resulting in high density. On the other hand, acidic functional group could be introduced to the carbon surface of the CAC by oxidation during the activated process which will induce polarity on the surface of the CAC [15,16].

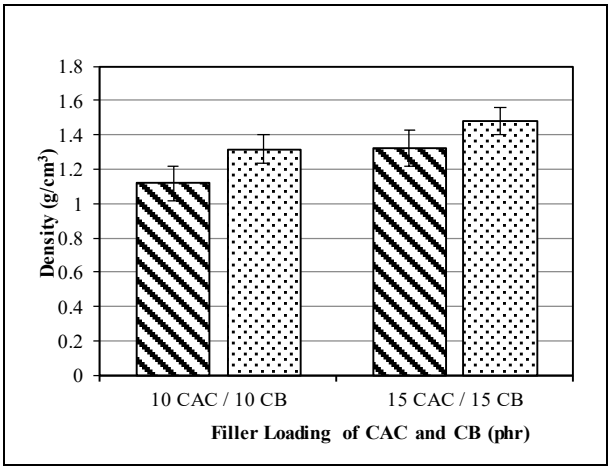

Figure 1 Density of 10 phr and 15 phr of CAC and CB in NR

Hardness

Hardness is sensitive to filler loading; thus, it is the most widely used test in the rubber industry, particularly in quality control. According to Phanny et al. [17], indentation hardness is inversely related to penetration and is dependent on the material's viscoelastic behaviour and elastic modulus. Figure 2 shows the hardness values for 10 phr and 15 phr of CAC and CB in NR. Increased CAC loading from 10 phr to 15 phr resulted in 40% increment in the hardness value. While the hardness for CB was higher than CAC at both loading, the increment of CB loading from 10 phr to 15 phr shows no significant changes. It is possible to conclude that there is an uneven distribution of CB in the rubber compound, which reduced the capability of the CB to increase the hardness [18]. The stiffness of the compound influenced the hardness. When the elasticity of rubber is reduced, the rigidity of the rubber compound increases because the stiffness of the rubber compound increases. This increment hardness value with increasing CAC loading demonstrated that adding CAC loading to the NR matrix results in a linear increase in material hardness. It can be said that, in order for the CAC to achieve the hardness value showed by 10 phr of CB, about 15 phr or more is needed.

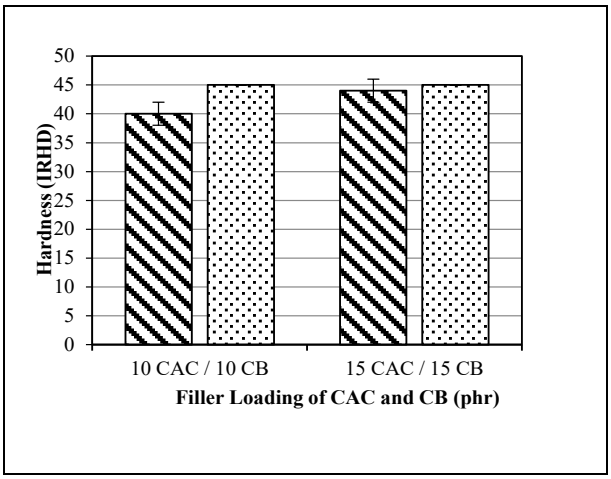

Figure 2 Hardness (IRHD) of 10 phr and 15 phr of CAC and CB in NR

Swelling Index (%)

The swelling index (%) of 10 phr and 15 phr of CAC/NR and CB/NR were shown in Figure 3. A significant different in swelling index (%) of CAC/NR compared to CB/NR can be seen from the figure whereas CAC possessed higher swelling index (%) than CB for both filler loading. The higher swelling ratio of CAC was possibly due to the original channel and porous structure of CAC which assisting the penetration of toluene solution into the filled NR. In addition, there is no significant changes in the swelling index (%) with the increased CAC loading from 10 phr to 15 phr. The swelling index (%) for CB/NR, on the other hand, showed 16% decrement as the CB loading increased. It is possible to said that the increasing loading of CB resulted in increased of stiffness and reduction in the flexibility which also limit the molecular movement of the rubber. Hence, the penetration of the toluene into the crosslinked structure of CB/NR will be difficult, thus lowering the swelling index (%) [19]. Saware [20] also stated that an equilibrium absorption in organic solvent is influenced by the degree of crosslink density, filler dispersion, filler nature, and solvent. As a result, more barriers are formed to the diffusing molecules in a well-crosslinking material, lowering the amount of the penetrated.

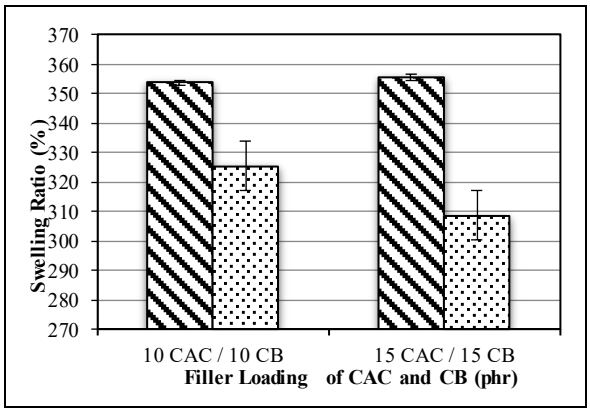

Figure 3 Swelling Index (%) of 10 phr and 15 phr of CAC and CB in NR

Mechanical Properties

Tensile Properties

Tensile strength, modulus at 100% (M100), modulus at 300% (M300), and elongation at break were listed in Table 3, while Figure 4 to 7 illustrated the tensile values in graphs.

Table 3 Data result for tensile test of rubber compound

Samples	Tensile strength (MPa)	Modulus M100 (MPa)	Modulus M300 (MPa)	Elongation at Break (%)
10 CAC	7.12	1.48	5.57	395.35
15 CAC	11.31	1.90	9.38	332.40
10 CB	17.04	1.82	9.67	393.75
15 CB	16.07	2.16	12.92	338.23

According to the Figure 4, the addition of CAC from 10 phr to 15 phr had increased the tensile strength. On the other hand, the tensile strength of CB/NR slightly reduced as the filler increased from 10 phr to 15 phr. This might be related to the uneven distribution of CB as previously mentioned. Phanny et al. [17] also discovered that the tensile strength of a CB-filled NR compound rises to a certain CB loading and then falls at higher loading due to the CB dispersion effect. This was due to bulk agglomeration caused by higher filler-filler interaction than filler-matrix interaction. Despite of the slightly reduced, the values of tensile strength for CB/NR were higher than the CAC/NR for both loading.

Mechanical properties strongly depend on the degree of surface interaction or compatibility between the filler and the vicinity rubber. The possibility of activated carbon to be slightly polar due to the oxidation during the activated process might influence the degree of compatibility of CAC with rubber matrix. Despite of the increased tensile strength at 15 phr of CAC, the value is still considered lower than the tensile strength of 10 phr of CB. Good compatibility between non-polar CB and

non-polar rubber matrix might contribute to this behavior. The excellent frame structure could support the stress transferred from the matrix and effectively improve the tensile strength of the rubber composite due to the better filler-rubber interaction. The results obtained agreed with Meng et al. [21] which discovered that SBR materials containing bamboo charcoal powder had a lower tensile strength than those containing carbon black. According to Govindan [22], CB filled rubber has nearly ten times the reinforcing potential of raw rubber, indicating the inherent reinforcing potential of CB.

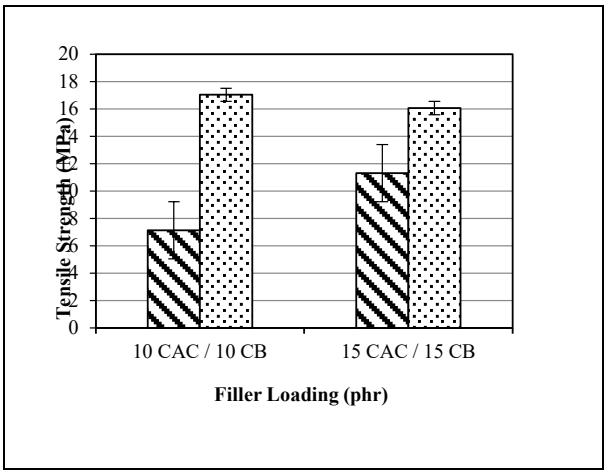

Figure 4 Tensile Strength of 10 phr and 15 phr of CAC and CB of NR

According to the Figure 5, the percentage of elongation at break decreased as CAC and CB content increased from 10 phr to 15 phr. This result might also contribute by the weak CAC-rubber interaction and uneven distribution of CB. Rubber have a good ability to significantly change their size and shape at low elongation by adjusting their conformation through backbone bond rotation [17]. Increased filler loading tends to limit the flexibility and movement of the rubber chains, causing vulcanization to fail at a lower elongation.

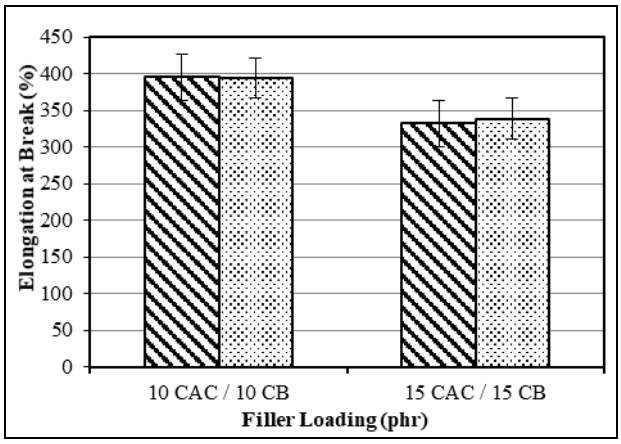

Figure 5 Elongation at Break of 10 phr and 15 phr of CAC and CB of NR

Meng et al. [21] discovered that as the loading of bamboo charcoal powder increased, elongation at break increased until it reached a maximum at 20 phr and then decreased. Other than that, the decrease in elongation at break was caused by the rubber composite's increasing stiffness and brittleness, as indicated by the increased in the hardness values and also the M300. Muniandy et al. [23] discovered that natural rubber/rattan powder/carbon black composites had the lowest elongation at break when compared to NR/RP/mica and NR/RP/$CaCO_3$. This could be due to a lower rubber-filler interaction, which causes filler agglomeration and reduces elongation at break. According to Mohamed et al. [24], the modulus is theoretically inversely proportional to the elongation at break. Due to the CB reducing the elasticity, the elongation at break decreases as the CB increases between 20 and 45 phr.

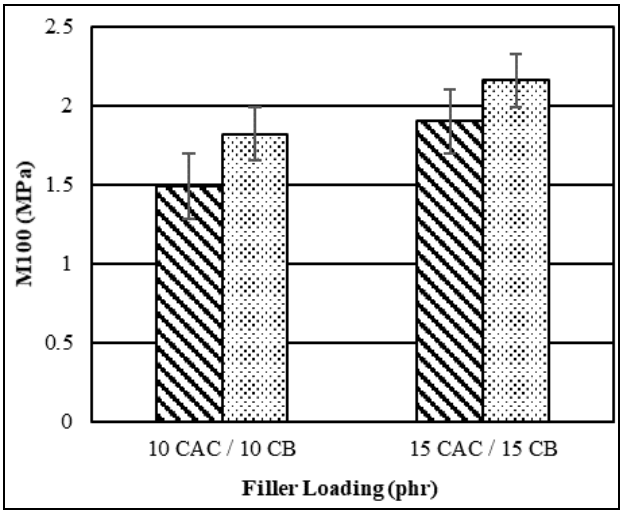

Figure 6 Modulus M100 of 10 phr and 15 phr of CAC and CB of NR

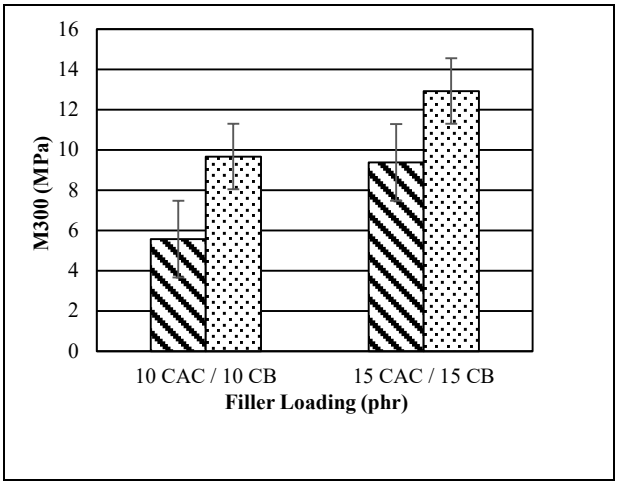

Figure 7 Modulus M300 of 10 phr and 15 phr of CAC and CB of NR

Figure 6 and 7 show the modulus of 100% (M100) and modulus at 300% (M300). Both samples showed increasing pattern of M100 and M300 as the filler loading increased, where CB/NR always exhibited higher modulus than CAC/NR. Mohamed et al. [24] also discovered that at 300% elongation, the tensile modulus increases with increasing CB loading but decreases at 50 phr because of filler aggregation. As more filler is used, agglomeration of the filler occurs. As a result, non-uniform dispersion of filler particulates occurred, resulting in

the formation of stress concentration points. Poor interfacial adhesion with the rubber matrix was also present as the amount of CAC increased, owing to an increase in agglomeration of CAC in the rubber matrix. The increase in tensile modulus was also associated with an increase in the MH value of the composites, which increased with filler loading [10]. Rodgerd et al. [25] also stated that the 300% modulus of saponified natural rubber composites increased with increasing macca charcoal content. However, as the loading of CB increased, the tensile strength began to decline. This is because the increased amount of CB caused agglomeration between the carbon black particles resulting in a reduction in interfacial interaction [24].

4.0 CONCLUSION

In conclusion, the CAC has a potential to be used as reinforcement filler in elastomers due to the porous structure which supposedly can provide more surface area for interaction with vicinity matrix. However, the loading of CAC needed in order to compete with CB is higher, whereas the tensile strength of 15 phr of CAC was still 34 % lower than 10 phr of CB. The density of CAC/NR was lower, and the swelling index was higher compared to the CB/NR which contributed by the porous structure of CAC. The porosity of the CAC will create void which reduce the density and also indirectly providing a space for the penetration of toluene since the texture of the filled rubber is not compact. Despite of that, the hardness, tensile strength, M100 and M300 of the CAC/NR showed a promising enhancement with the increment in filler loading from 10 phr to 15 phr.

Acknowledgement

The authors would like to relay our gratitude to the Faculty of Applied Sciences, Universiti Teknologi MARA, Arau Campus and Shah Alam Campus for the facilities and financial provided.

References

[1] Xue, B., Wang, X., Sui, J., Xu, D., Zhu, Y. and Liu, X. 2019. A Facile Ball Milling Method to Produce Sustainable Pyrolytic Rice Husk Bio-Filler for Reinforcement of Rubber Mechanical Property. *Industrial Crops and Products*. 141: 11179. DOI: https://doi.org/10.1016/j.indcrop.2019.111791

[2] Mark, J. E., Erman, B. and Roland, M. 2015. Science and Technology of Rubber, 4th ed. Elsevier, 2005. ISBN: 978-0-12-394584-6

[3] Kohjiya, S. and Ikeda, Y. 2021. Chemistry, Manufacture and Applications of Natural Rubber, 2nd ed. Woodhead Publishing Series in Materials, Elsevier. ISBN: 9780128188446

[4] Cindy, S. B. and Katrina. C. 2016. High Performance Waste-Derived Filler/Carbon Black Reinforced Guayule Natural Rubber Composites. *Industrial Crops and Products*. 86: 132-142. DOI: https://doi.org/10.1016/j.indcrop.2016.03.021

[5] Thaptong, P., Sirisinha, C., Thepsuwan, U. and Sae, O.P. 2014. Properties of Natural Rubber Reinforced by Carbon Black-based Hybrid Fillers. *Polymer - Plastics Technology and Engineering.* 53(8): 818–82.3. DOI:. https://doi.org/10.1080/03602559.2014.886047

[6] Sattayanurak, S., Noordermeer, J. W. M., Sahakaro, K., Kaewsakul, W., Dierkes, W. K. and Blume, A. 2019. Silica-Reinforced Natural Rubber: Synergistic Effects by Addition of Small Amounts of Secondary Fillers to Silica-Reinforced Natural Rubber Tire Tread Compounds. *Advances in Materials Science and Engineering.* 1-8. DOI: https://doi.org/10.1155/2019/5891051

[7] Chen, W. J., Gu, J. and Xu, S. H. 2014. Exploring Nanocrystalline Cellulose as a Green Alternative of Carbon Black in Natural Rubber / Butadiene Rubber / Styrene-Butadiene Rubber Blends. *Express Polymer Letters.* 8 (9): 659–668. DOI: 10.3144/expresspolymlett.2014.69

[8] Alkhazraji, A. N. 2018. Enhancement of Mechanical Properties and Handling Characteristic of Tire Rubber Using Different Percentage of Nano Aluminum Oxide and Carbon Black. *International Journal of Nanoelectronics and Materials.*11 (3): 263-270. DOI:http://dspace.unimap.edu.my:80/xmlui/handle/123456789/57581

[9] Omnes, B., Thuillier, S., Pilvin, P., Grohens, Y. and Gillet, S. 2008. Effective Properties of Carbon Black Filled Natural Rubber: Experiments and Modeling. Elsevier - Composites: Part A: Applied Science and Manufacturing. 39 (7): 1141-1149. DOI: https://doi.org/10.1016/j.compositesa.2008.04.003

[10] Meng, X., Zhang, Y., Lu, J., Zhang, Z., Liu, L. and Chu, P. K. 2013. Effect of Bamboo Charcoal Powder on the Curing Characteristics, Mechanical Properties, and Thermal Properties of Styrene-Butadiene Rubber with Bamboo Charcoal Powder. *Journal of Applied Polymer* Science. 130 (6): 4534–4541. DOI: https://doi.org/10.1002/app.39522

[11] Fatin, M.H., Zulkepli, N. N., Kamarudin, H., Zakaria, M. S., Rohaizad, N. M., Ismail, H., Abdullah, M. M. A. B. and Sam, S.T. 2015. Characteristics and Physical Properties of Imperata Cylindrica Activated Carbon Filled SMR L Compounding. *Applied Mechanics and Materials*, 815: 44-48. DOI: 10.4028/www.scientific.net/AMM.815.44.

[12] Chigondo, F., Shoko, P., Nyamunda, B. C., Guyo, U. and Moyo, M. 2013. Maize Stalk as Reinforcement in Natural Rubber Composites. *International Journal of Scientific & Technology Research.* 2 (6): 263-271. DOI: http://hdl.handle.net/11408/1006

[13] Ahmed, K. 2015. Hybrid Composites Prepared from Industrial Waste: Mechanical and Swelling Behaviour. *Journal of Advance Research*. 6: 225-232. DOI*:* https://doi.org/10.1016/j.jare.2013.12.002

[14] Zhu, S., Guo, Y., Chen, Y., Su, N., Zhang, K. and Liu, S. 2016. Effects of the Incorporation of Nano-Bamboo Charcoal on the Mechanical Properties and Thermal Behavior of Bamboo-Plastic Composites. *Bioresources.* 11(1): 2684–2697. DOI*:* 10.15376/biores.11.1.2684-2697

[15] Shafeeyan, M. S., Daud, W. A., Houshmand, A. and Shamiri, A., J. 2010. A Review on Surface Modification of Activated Carbon for Carbon Dioxide Adsorption. *Journal of Analytical and Applied Pyrolysis.* 89: 143-151. DOI*:* http://dx.doi.org/10.1016/j.jaap.2010.07.006

[16] Chen, J., Ma, Q.C and Chen Q. (2012). Influence of Surface Functionalization via Chemical Oxidation on the Properties of Carbon Nanotubes. J. Colloid Interface Sci. 370: 32–38. DOI : https://doi.org/10.1016/j.jcis.2011.12.073

[17] Phanny, Y., Azura, A. R. and Ismail, H. 2012. Effect of Different Origins of Natural Rubber on the Properties of Carbon Black Filled Natural Rubber Composites. *ASEAN Engineering Journal Part B.* 2 (1): 60–67. Retrieved from https://eprints.usm.my/43116/1/Pages_from_EFFECT_OF_DIFFERENT_ORIGIN.pdf

[18] Ichazo, M.N., Albano, C., Hernandez, M., Gonzalez, J. and Carta, A. 2008. Effects of Particle Size and Size Distribution on the Mechanical Properties of EPDM/Silica Vulcanizates. *Advanced Materials Research.* 47-50: 113-116. DOI: 10.4028/www.scientific.net/AMR.47-50.113

[19] Wahab, N. M. A., Azman, A., Majid, N. A., Ismail, S. N. S. and Mustafa, M. S. 2019. Effect of Marble Tile Waste Powder (MTWP) Filler on Cure and Mechanical Behaviour of Natural Rubber (NR) Compound. *Jurnal Intelek*. 14 (2):220-229. DOI : 10.24191/ji.v14i2.238

[20] Sawere, B. T. 2016. Effect of Egg Shell and Rice Husk on Hardness and Swollen Properties of Natural Rubber Vulcanizates. *International Journal of Advancements in Research & Technology*. 5(8): 36–43. Retrieved from http://www.ijoart.org/docs/EFFECT-OF--EGG-SHELL-AND-RICE-HUSK-ON-HARDNESS-AND-SWOLLEN-PROPERTIES-OF-NATURAL-RUBBER-VULCANIZATES.pdf

[21] [21] Meng, X., Zhang, Y., Lu, J., Zhang, Z., Liu, L. and Chu, P. K. 2013. Effect of Bamboo Charcoal Powder on the Curing Characteristics, Mechanical Properties, and Thermal Properties of Styrene-Butadiene Rubber with Bamboo Charcoal Powder. *Journal of Applied Polymer Science*. 130 (6): 4534–4541. DOI: https://doi.org/10.1002/app.39522

[22] Govindan, T. 2018. Comparative Studies on the Mechanical Properties of Natural Rubber and Natural Rubber filled with Multi-walled Carbon Nanotubes. *AIP Conference Proceeding*. 1963, 020009. DOI : https://doi.org/10.1063/1.5036855

[23] Muniandy, K., Ismail, H. and Othman, N. 2012. Effects of Partial Replacement of Rattan Powder by Commercial Fillers on the Properties of Natural Rubber Composites. *BioResources.* 7 (4): 4640–4657. DOI: 10.15376/biores.7.4.4640-4657, Science & Electronics, 13(2): 48–52, 2015. Retrieved from 10.1063/1.5036855

[24] Mohamed, R., Nurazzi, N. M. and Huzaifah, M. 2017. Effect of Carbon Black Composition with Sludge Palm Oil on the Curing Characteristic and Mechanical Properties of Natural Rubber/Styrene Butadiene Rubber Compound. *IOP Conference Series: Materials Science and Engineering.* 223 (1), 012008. DOI: 10.1088/1757-899X/223/1/012008

[25] Rodgerd, P., Yakkul, S. and Sakdapipanich, J. 2013. Application of Macca Charcoal in Saponified Natural Rubber Composites. *Advanced Materials Research.* 844: 149–153. DOI: https://doi.org/10.4028/www.scientific.net/AMR.844.149