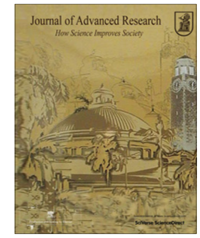
Journal of Advanced Research (2015) 6, 225–232

Cairo University

Journal of Advanced Research



ORIGINAL ARTICLE

Hybrid composites prepared from Industrial waste: Mechanical and swelling behavior

Khalil Ahmed *

Applied Chemistry Research Centre, PCSIR Laboratories Complex, Karachi 75280, Pakistan

ARTICLE INFO

Article history:
Received 16 June 2013
Received in revised form 3 December 2013
Accepted 4 December 2013
Available online 11 December 2013

Keywords:
Marble waste powder
Hybrid composite
Carbon black
Silica
Mechanical properties

ABSTRACT

In this assessment, hybrid composites were prepared from the combination of industrial waste, as marble waste powder (MWP) with conventional fillers, carbon black (CB) as well as silica as reinforcing material, incorporated with natural rubber (NR). The properties studied were curing, mechanical and swelling behavior. Assimilation of CB as well as silica into MWP containing NR compound responded in decreasing the scorch time and cure time besides increasing in the torque. Additionally, increasing the CB and silica in their respective NR hybrid composite increases the tensile, tear, modulus, hardness, and cross-link density, but decreases the elongation and swelling coefficient. The degradation property e.g., thermal aging of the hybrid composite was also estimated. The overall behavior at 70 °C aging temperature signified that the replacement of MS by CB and silica improved the aging performance.

Introduction

Pollution that is created as the outcome of human activities has banged onto the environment that life is about to face unexpected calamity. At present, there are hundreds of tones of gases, liquid and solid industrial wastes that spoil the soil, water and atmospheric environment and have unhealthy effect on human.

A gigantic quantity of marble waste is generated in marble carving industry as a by-product during the cutting/polishing process of marble blocks and is carried away by the drainage system or thrown away on open grounds. Consequently, employing of marble waste in the fabrication of new substances will assist to defend the environment. Polymer composites can be the best application to utilize marble sludge waste in large quantities to substitute the conventional fillers, clay and other materials. A quite few attempts have been made to use marble waste in road making, soil filling, and building construction materials [1] and asphaltic concrete [2] but very little effort together with our own have been made to employ as filler in rubber composites [3–6].

The polymer based composites, act as matrix while carbon black, silica and clay are act as reinforcing material.

Fillers enhance the mechanical properties for example, tensile, tear, hardness and abrasion resistance of the final product. Carbon Black and silica usually plays the vital role as reinforcing material of the rubber compounding and can also reduce the production cost [7–12]. Nowadays, mineral filler also added to polymers to achieve the improved product with low cost [13–20]. It is well-known that for cured filled rubber, the worth of reinforcement depends on the interaction of various

* Tel.: +92 21 34690350; fax: +92 21 34641847.
E-mail address: khalilmsrc@gmail.com

Peer review under responsibility of Cairo University.

ELSEVIER Production and hosting by Elsevier

2090-1232 © 2013 Production and hosting by Elsevier B.V. on behalf of Cairo University.
http://dx.doi.org/10.1016/j.jare.2013.12.002

filler related considerations counting the particle size, shape, dispersion, surface area, surface reactivity, structure of the filler, and bonding quality between the fillers and the matrix.

Currently different hybrid filler synchronization systems have been examined by numerous researchers. CB–silica hybrid filler system glances to be the most popular and successful. The dual phase filler is at present commercially manufactured by Cabot Corporation for the applications of truck tire [21]. The CB–silica hybrid filler system recommends generally overall improved mechanical properties compared to individual one. It also illustrates the most favorable balance of various properties for example wet traction, wear resistance and rolling resistance [22].

Rattanasom et al. [23] used the CB/silica hybrid filler with natural rubber and found better overall mechanical properties at 30/20 and 20/30 hybrid filler ratio. Besides that, RNP/carbon black and RNP/silica hybrid [24] and carbon black/nanoclay [25], carbon black/clay [26] have also been evaluated as reinforced hybrid systems.

Another low cost composite is carbon black and silica amended with newspaper (recycled). The use of combined fillers in polymers has been recognized for many years [27,28] persuaded by the demand for high performance materials.

Herein the study, the effects of partial or full replacement of marble waste by carbon black as well as silica, in hybrid composite on the overall properties is reported. Tensile, tear strength, modulus, elongation at break and hardness was investigated moreover swelling properties was also carried out to calculate the swelling ratio, crosslink density and shear modulus of hybrid filled NR composites.

Experimental

Materials

The raw elastomer used was a natural rubber, NR (grade RSS-1) poly cis-isoprene, supplied by Rainbow rubber industry. The conventional reinforcing filler was carbon black (N330, particle 40–50 nm, specific gravity 1.80–1.82). Precipitated silica (Zeosil-175) by Rhodia. Marble waste as a sludge form, was obtained from home marble industry.

The MWP was dehydrated in the oven and then pulverized in finer form and passed through sieve to obtain 37 μm.

In addition to rubber, filler and other components such as Tetramethyl thiuram disulfide (TMTD), zinc oxide, sulfur as curing agent, 3-Dimethylbutyl-N-phenyl-p-phenylenediamine as antioxidant, were used as commercial grade and procured from the market.

Characterization of MWP by XRF spectrometer

The characterization of MWP was carried out with a XRF spectrometer (PIONEER with the Bruker AXS SPECTRA).

Preparation of composites

The basic formulation is given in Table 1. The composites were prepared as described previously [5,6,29].

Cure characteristics

The cure characteristics of mixes were studied as in our earlier works [30,31].

Testing of physical properties

Testing of mechanical, swelling and aging properties of MWP/CB and MWP/Silica hybrid NR composites were carried out as described previously using standard procedures [4,32].

Results and discussion

Characterization of marble waste powder

The chemical composition of MWP was analyzed by X-ray fluorescence spectrometer (Bruker AXS, Germany). The chemical examining of MWP showed the occurrence of Calcium Oxide (68.6%), Magnesium Oxide (22.13%) as main components besides with Silica (3.89%), Aluminum Oxide (2.785%), Iron Oxide (0.603%), Chromium Oxide (0.24%) Zinc Oxide (0.20%) and Titanium Oxide (0.549%).

Obviously the composition of MWP shows calcium and magnesium compound in large amount. Silica, aluminum oxide and iron oxide also present in small amount.

Curing characteristics

The values of curing characteristics were determined from the corresponding curing isotherms measured at 155 °C. Figs. 1

Table 1 Compound formulation of the constructed hybrid NR composites.

Component	Part per hundred of rubber (phr)
NR	100
ZnO	05
Stearic acid	02
TMTD[a]	2.4
Antioxidant[b]	1.5
Sulfur	1.6
Marble waste[c]/carbon black	00/00, 60/00, 50/10,40/20, 30/30, 20/40, 10/50, 00/60
Marble waste[c]/silica	00/00, 60/00, 50/10,40/20, 30/30, 20/40, 10/50, 00/60
Si-69	1.2

[a] Tetra methylthiuram disulfide.
[b] 3-Dimethylbutyl-N-phenyl-p-phenylenediami.
[c] MWP particle size, 37 μm.

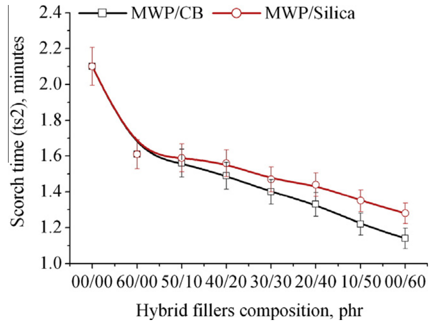


Fig. 1 Scorch time of the hybrid composites.

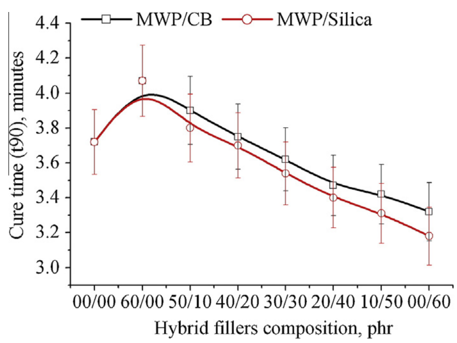


Fig. 2 Cure time of the hybrid composites.

and 2 shows the assessment of replacement of MWP by CB as well as silica on scorch and cure time of the hybrid composite. The result illustrates that the scorch time as well as cure time reduced by increasing the amount of CB as well as silica in their particular hybrid system as compare to the unfilled and 60 phr MWP in NR composites. The high energy and heat develops during compounding, owing to greater viscosity with better shear heating of the rubber compounds results in diminishing scorch time as well as cure time. Reduction in scorch time by increasing CB as well as silica was suitable to develop additional cross-linked in the MWP/CB and MWP/silica dual filler system.

The minimum and maximum torque increases by increasing CB as well as silica amount in their respective hybrid NR composite that is shown in Figs. 3 and 4. The torque generally lied on the degree of cross link and strength of the filler particles

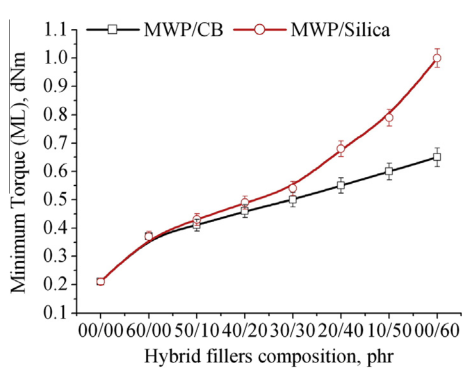


Fig. 3 Minimum torque of the hybrid composites.

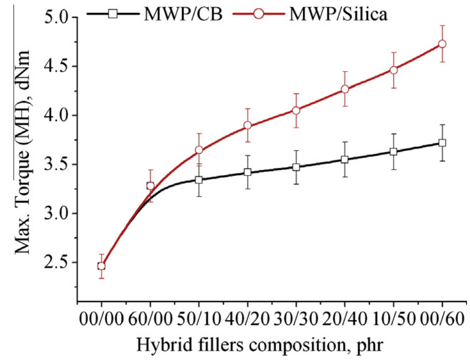


Fig. 4 Maximum torque of the hybrid composites.

within the matrix [33] which trim downs the molecular manipulability of the matrix chain, moreover the result in rigid construction of the composite. Enhancement in the torque proved that continuing insertion of CB and silica in their corresponding NR hybrid composite developed the crosslink density of the NR compounds.

CB as well as silica is reinforcing fillers with having smaller particle and so much better interaction between the filler and NR as compare to MWP.

From obtained data it is clear that the addition of CB as well as silica in MWP with its content in NR compound influences the properties of prepared hybrid composites in various ways.

The CRI result given in Fig. 5 illustrates no considerable outcome on the ratio of CB and silica in NR composite.

Mechanical properties

The stress–strain curves of MWP/CB and MWP/silica hybrid composite were evaluated and results are shown in Figs. 6 and 7. From Figs. it is clear that the stiffness as governed by the slope of the initial linear part increases with increasing the concentration of CB and silica that acts as reinforcing filler in MWP/CB and MWP/silica hybrid composites. Consequently, elongation at break decreases. The stress–strain parameters; modulus of the samples as realized from the stress–strain curves were found to be significantly affected by increasing CB or silica concentration. Both results show that CB as well as silica content dependence of these parameters. Generally, it is clear that modulus increases with increasing

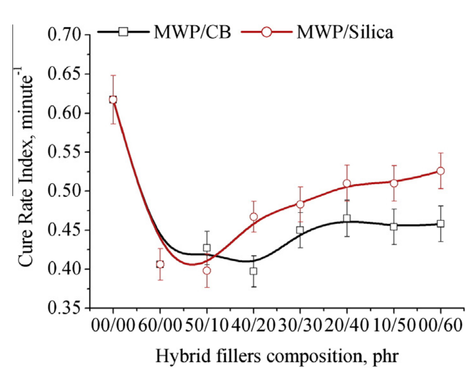


Fig. 5 Cure rate index of the hybrid composites.

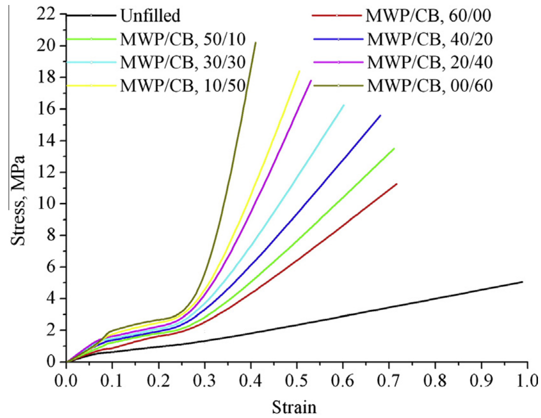


Fig. 6 Stress–strain curves of the MWP/CB hybrid composites.

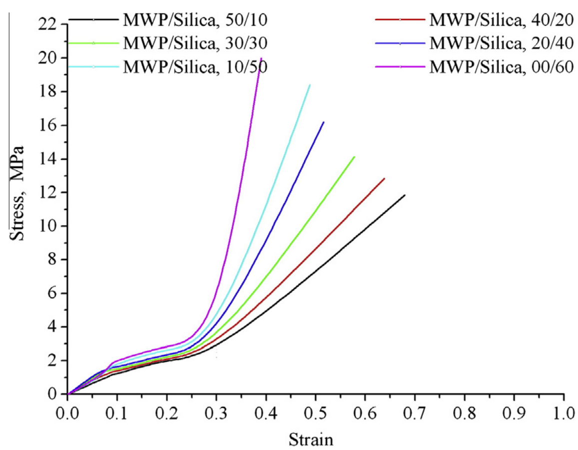


Fig. 7 Stress–strain curves of the MWP/Silica hybrid composites.

CB and silica content and reaches a maximum value at 60 phr of CB and silica amount in their particular NR composites. On the other hand, strain shows a minimum value around the same content. The consequent formation of compact structure [34] with increasing the contents of CB or silica may be conscientious for the minimum in strain and the maximum of stress observed at 60 phr of CB or silica amount.

The tensile strength of hybrid composites, before and after aging is revealed in Table 2. As expected, incorporation of CB as well as silica replaced by MWP in their relevant hybrid NR composite has improved tensile strength. This improvement was owing to the adequate interaction of carbon black as well as silica with NR over MWP. The well-built filler–rubber interaction increased the ability of stress to shuffle from matrix to filler therefore, boosted the tensile strength.

This is also attributed to good filler dispersion in the NR, inter-tubular and interfacial interactions of the filler and rubber besides to the filler intercalation with rubber constituents.

The filler, reinforcing level improves with reduce particle size or besides extend in surface area. Additional proficient will be stress shift from matrix to filler, when diminish the particle size [35].

The same tensile trend is viewed in samples after aging. However, a decrease is found in all MWP/CB and MWP/silica hybrid composite [36]. Thermal aging of MWP/CB and MWP/silica hybrid composite caused the tensile to be worse, particularly 100 °C for 96 h [37]. Both aging properties show less than 100% retention though, aging at 70 °C of hybrid composite observe higher retention value of tensile as compare to 100 °C. This is due to better thermal stability at lower aging temperature.

According to the results, the tensile strength is much higher for the marble sludge/carbon black-filled NR hybrid composite than for the marble sludge/silica filled hybrid ones. This may be explaining as the reinforcing level and the crosslink density of both different types of filler. The reinforcing altitude is improved for the better dispersion of filler in matrix. Silica is poorer dispersion as compare to carbon black because silica has strong filler–filler interaction by silanol groups. The tensile strength of unfilled NR compound was not broken. Non-failure specimen has less peak load and higher elongation at break. However, when hybrid composites prepared the tensile strength gradually increases and the failure behavior raised with higher peak load and lower the strain.

Table 3 shows the 200% modulus (before and after aging) of MWP/CB and MWP/silica hybrid composite. The result shows that the highest value of modulus for hybrid composite was obtained. Progressive substitution of MWP assisted better dispersion of CB as well as silica within matrix. It is observed that the modulus of MWP/CB and MWP/silica hybrid composites improved with the increasing CB as well as silica in their individual hybrid composite. This is due to the small particle size, CB as well as silica and their aptitude to have excellent dispersion within the matrix as compare to MWP that increased the rigidity of composites. Higher retention value have been revealed at 70 °C, of 200% modulus of hybrid composite that may be due to the post curing and cross-linking, but at 100 °C the% retention is less than 100 in 200% modulus is shown.

The modulus at 90–110 °C decreases, conversely at 70–90 °C the pace of modulus increase due to the intricacy of reactions taking place in vulcanized rubber [38,39].

Clarke et al. [40] investigated the thermal aging kinetics of NR compound and point up that cross-linking reaction and scission reaction ascend the rate of reaction through increase in aging temperature. The scission reactions have greater activation energy as compare to crosslink reaction. Therefore, with decrease in aging temperature, the rate of scission decreases at 70–80 °C. The rate of crosslink in fact increases when aging temperature decreases.

Table 4 represents the tear strength trend of MWP/CB and MWP/silica hybrid composite. The tear strength also shows similar as to tensile strength. Tear strength of filled hybrid composite is higher than that of the unfilled as well as 60 phr of MWP filled NR composite. With addition of CB as well as silica in the particular hybrid system, the tear stress equivalently dispersed in NR and hybrid composite shows enhanced in tear strength.

Table 5 summarizes the% elongation at break of MWP hybrid system through CB as well as silica NR composite with accelerated thermal aging. When CB as well as silica is added in-place of MWP, the elongation at break decreased. This reduction indicates that ductility became worse when more conventional fillers were added into NR composites. The CB and silica are non-deformable particle so the addition of CB as well as silica in their particular composites restricts the molecular chains movement.

Table 2 Summary of tensile strength for MWP/CB and MWP/silica hybrid system filled NR composite.

Hybrid filler composition	Tensile strength (MPa)					
	MWP/CB			MWP/silica		
	Before aging	After aging at 70 °C	After aging at 100 °C	Before aging	After aging at 70 °C	After aging at 100 °C
00/00	5.05	4.10	1.97	–	–	–
60/00	11.25	9.43	4.65	11.25	9.23	4.65
50/10	13.50	12.05	5.73	11.86	10.91	5.65
40/20	15.60	14.03	6.70	12.85	11.86	5.84
30/30	16.24	14.68	7.00	14.15	13.26	6.15
20/40	17.80	16.20	8.00	16.20	14.97	7.10
10/50	18.38	16.86	8.80	18.40	17.10	8.12
00/60	20.20	18.58	9.92	20.00	18.53	9.10

Table 3 Summary of 200% Modulus for MWP/CB and MWP/silica hybrid system filled NR composite.

Hybrid filler composition	200% Modulus (MPa)					
	MWP/CB			MWP/silica		
	before aging	After aging at 70 °C	After aging at 100 °C	Before aging	After aging at 70 °C	After aging at 100 °C
00/00	1.00	1.21	0.72	–	–	–
60/00	1.77	2.26	1.53	1.77	2.26	1.53
50/10	1.92	2.47	1.69	2.13	2.52	1.85
40/20	1.98	2.55	1.74	2.19	2.60	1.95
30/30	2.14	2.84	1.93	2.30	2.80	2.08
20/40	2.29	3.10	2.10	2.41	3.03	2.22
10/50	2.56	3.30	2.37	2.74	3.49	2.62
00/60	2.78	2.87	2.58	2.94	3.88	2.84

Table 4 Summary of tear strength for MWP/CB and MWP/silica hybrid system filled NR composite.

Hybrid filler composition	Tear strength (N/mm)					
	MWP/CB			MWP/silica		
	Before aging	After aging at 70 °C	After aging at 100 °C	Before aging	After aging at 70 °C	After aging at 100 °C
00/00	13.60	10.30	08.32	–	–	–
60/00	21.46	16.90	13.95	21.46	16.90	13.95
50/10	25.36	20.61	14.20	23.19	17.54	12.20
40/20	26.12	22.00	14.97	24.40	20.45	13.15
30/30	29.86	25.68	17.47	27.11	23.26	15.08
20/40	34.50	30.40	20.63	30.17	26.54	17.38
10/50	40.17	35.90	24.46	34.81	30.60	20.23
00/60	44.60	41.10	27.80	39.00	35.88	23.15

The decrease in the elongation at break also due to the striking forces between the filler and the polymer molecules leading to the development of a cross-linked structure so as to limit the free mobility of the polymer chains, hence increases the resistance to accelerate upon the execution of tension [41].

Table 6 depicts the variation in hardness of correspondingly MWP/CB and MWP/silica hybrid composite throughout accelerated thermal aging. It is evident that hardness of corresponding composite increased via replacement of MWP by CB as well as silica. This discovery is in well conformity with 200% modulus outcomes cited prior in this research works that disclosed an increase in rigidity of the composites with addition of CB as well as silica within hybrid composites.

However, the MWP/silica hybrid composite has relatively higher hardness as compared to MWP/CB hybrid composite due to the two dissimilar filler with different form and dimensions which design a new filler establishment. After aging data of hardness of all the samples changed to some extent.

Swelling properties

Figs. 8 and 9 show the relationship of swelling ratio of toluene uptake at seven days (168 h) by MWP/CB and MWP/silica hybrid composite. It is clear from the figure that MWP/CB and MWP/silica filled system illustrates reduce solvent (toluene)

Table 5 Summary of% elongation at break for MWP/CB and MWP/silica hybrid system filled NR composite.

Hybrid filler composition	% Elongation at break					
	MWP/CB			MWP/silica		
	Before aging	After aging at 70 °C	After aging at 100 °C	Before aging	After aging at 70 °C	After aging at 100° ° C
00/00	988	665	553	–	–	–
60/00	716	497	374	716	497	374
50/10	710	505	383	680	480	354
40/20	681	502	378	638	471	336
30/30	601	450	348	578	417	311
20/40	530	407	314	516	390	288
10/50	505	396	307	488	386	279
00/60	410	328	254	390	325	232

Table 6 Summary of hardness for MWP/CB and MWP/silica hybrid system filled NR composite.

Hybrid filler composition	Hardness, shore-A					
	MWP/CB			MWP/silica		
	Before aging	After aging at 70 °C	After aging at 100 °C	Before aging	After aging at 70 °C	After aging at 100 °C
00/00	36.0	39.0	40.0	–	–	–
60/00	50.0	53.0	54.8	50.0	53.0	54.8
50/10	48.0	53.0	55.0	53.0	53.3	59.13
40/20	54.0	59.8	62.0	54.0	59.7	60.6
30/30	56.0	62.0	64.4	58.0	63.4	65.3
20/40	59.0	65.5	68.3	62.0	69.0	68.12
10/50	62.0	69.0	72.0	67.0	74.9	76.33
00/60	66.0	73.6	77.0	73.0	81.5	83.85

up-take propensity as contrasted with the unfilled. All MWP/CB and MWP/silica hybrid composites also displayed a similar pattern of sorption, where the filled NR composites absorbed solvent was very fast within the initial 36 h, followed by gradual increase until achieving a saturated point. Obviously, the toluene uptake of the NR composites decreased as CB or silica content increased in both hybrid systems. The efficient reinforcement in the hybrid NR composite systems is visible from the swelling ratio graphs. Vulcanization causes to limit the movements of the NR molecules in presence of fillers, although departs their fragmental mobility high [35,42]. Filler changes this state. The attachment of molecule chains to the filler has been achieved through sopping wet. CB and silica can wet rubber segments because the increased prospect of various absorptions.

The improvement of interfacial adhesion along with filler and matrix also reduced, toluene addition in interfacial spaces, therefore, limiting the penetration of toluene into the composites. The highest bound rubber is found with CB and silica particles in their particular hybrid NR composite systems. This reinforcement restricts the free movement of rubber chains, thus improves the toluene resistance.

Swelling coefficient (α) of MWP/CB as well as MWP/silica hybrid composites is presented in Fig. 10. From the diagram it is noticeable that, swelling coefficient of hybrid composites is decreased via replacement of MWP by CB as well as silica owing to better diffusion of CB as well as silica in rubber. However, some fragile interaction between silica and MWP/NR, the penetration of toluene into the composites is quite easier as compare to CB filled hybrid composite which has better interaction with NR than silica. Hence, the 'α' of CB containing hybrid composite reduced owing to the diffusion barrier of solvent (toluene) into the MWP/CB hybrid composite.

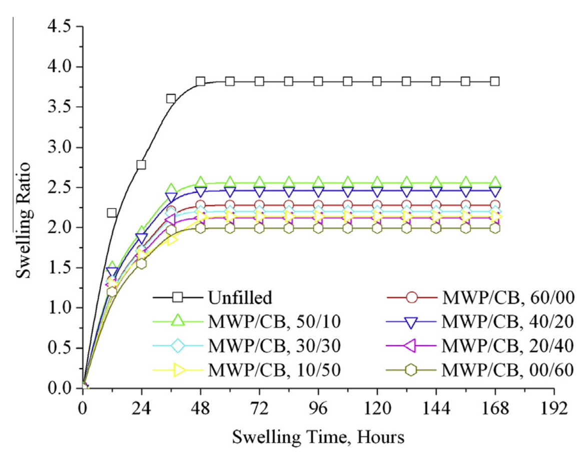


Fig. 8 Swelling ratio of the MWP/Silica hybrid composites.

The crosslink density data are illustrated in Fig. 11 that is increasing with gradual increasing amount of CB as well silica in MWP/CB and MWP/silica system.

The increase in filler-rubber interaction examined in MWP/CB and MWP/silica hybrid composites, due to increasing CB

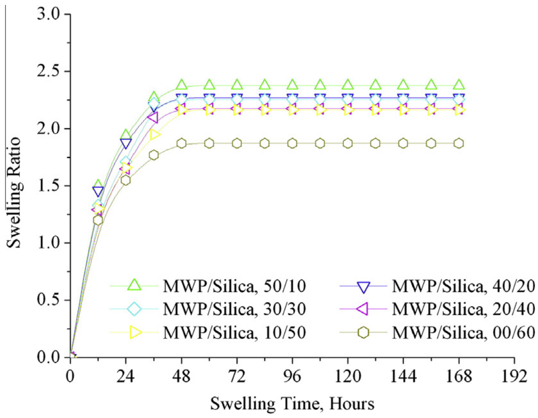


Fig. 9 Swelling ratio of the MWP/CB hybrid composites.

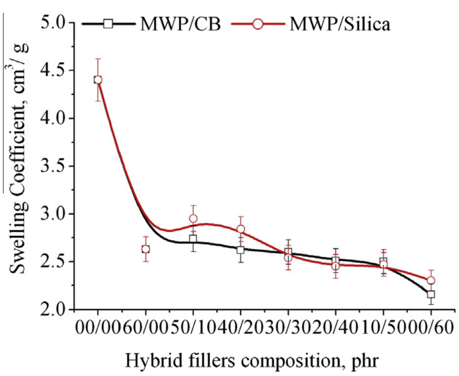


Fig. 10 Swelling Coefficient of the hybrid composites.

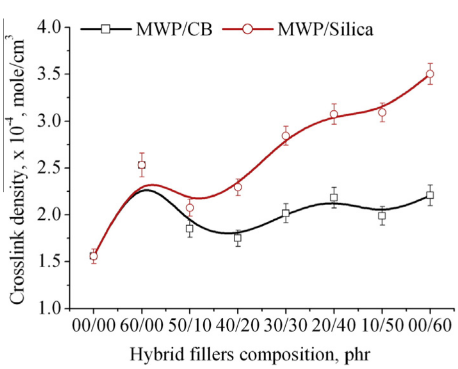


Fig. 11 Crosslink Density of the hybrid composites.

as well as silica content that has best surface activity and therefore high affinity of powerful interaction to rubber.

Conclusions

The partial replacement of industrial waste like MWP with conventional fillers as CB as well as silica in NR was investigated. The following conclusions have been depicted from the present assessments.

The cure time as well as scorch time decrease, but the torques increases with addition of CB as well as silica in the hybrid composite.

Remarkable mechanical performance, particularly tensile strength, tear strength, modulus and hardness (superior values), and% elongation at break (inferior values) were obtained with replacement of MWP by CB and silica. MWP/CB hybrid composite was higher than those with MWP/silica hybrid NR composites.

Addition of CB as well as silica hybrid composites had an effect on swelling ratio and cross link density of composites.

Low mechanical properties of the some MWP/CB and MWP/silica hybrid composite showed after thermal aging while modulus, hardness and swelling coefficient increased at 70 °C.

This assessment revealed that MWP can be utilized as a co-filler with CB as well as silica to obtain better dispersion of MWP/CB and MWP/silica hybrid filler with NR to gain, high mechanical properties.

Indentation results furthermore disclose that MWP can be utilized as a co-filler with CB as well as silica to make the better mechanical properties.

Conflict of interest

The authors has declared no conflict of interest.

Compliance with Ethics Requirements

This article does not contain any studies with human or animal subjects.

References

[1] Binici H, Hasan K, Salih Y. Influence of marble and limestone dusts as additives on some mechanical properties of concrete. Sci Res Essay 2007;2:372–9.
[2] Karasahin M, Terzi S. Evaluation of marble waste dust in the mixture of asphaltic concrete. Constr Build Mater 2007;21:616–20.
[3] Agrawal S, Mandot S, Bandyopadhyay S, Mukhopadhyay R. Use of marble waste in rubber industry: Part I (in NR compound). Prog Rubb Plast Recycl Technol 2004;20(3):229–46.
[4] Ahmed K. Properties of natural rubber composite incorporated with industrial waste/carbon black hybrid filler. Chem Lett 2013;42:1105–7.
[5] Ahmed K, Nizami SS, Raza NZ, Mahmood K. Characterization of mechanical properties of marble sludge/natural rubber composites. J Chem Soc Pak 2012;33:1468–76.
[6] Ahmed K, Nizami SS, Raza NZ. Reinforcement of natural rubber hybrid composites based on marble sludge/Silica and marble sludge/rice husk derived silica. J Adv Res 2014;5(2): 165–73.
[7] Park SM, Lim YW, Kim CH, Kim DJ, Moon WJ, Kim JH, et al. Effect of carbon nanotubes with different lengths on mechanical and electrical properties of silica-filled styrene butadiene rubber compounds. J Ind Eng Chem 2013;19:712–9.
[8] Wang M. The role of filler networking in dynamic properties of filled rubber. Rubb Chem Technol 1999;72:430–48.
[9] Hamed GR. Tear strength of black-filled natural rubber crosslinked via conventional. Efficient sulfur cures. Rubb Chem Technol 2004;77:227–9.
[10] Choi SS, Ha SH. Influence of the swelling temperature and acrylonitrile content of NBR on the water swelling behaviors of silica-filled NBR vulcanizates. J Ind Eng Chem 2009;15:167–70.

[11] Furtado CRG, Leblanc JL, Nunes RCR. Mica as additional filler in SBR–silica compounds. Eur Polym J 2000;36:1717–23.
[12] Arroyo AM, Lopez – Man A, Chado B. Organo-montmorillonite as Substitute of carbon black in natural rubber compounds. Polymer 2003;44:2447–53.
[13] Mohan TP, Kuriakose J, Kanny K. Effect of nanoclay reinforcement on structure, thermal and mechanical properties of NR/SBR J. Ind Eng Chem 2011;17:264–70.
[14] Da-Silva ALN, Rocha MCG, Movaces MAR, Valente CAR, Coutinho FM. Mechanical and rheological properties of composites based on polyolefin and mineral additives. Polym Test 2002;21:57–60.
[15] Helaly FM, El-sawy SM. Preparation and evaluation of some investigated natural and acrylonitrile rubber vulcanizates for physiotherapeutic purposes. Polym Plast Technol Eng 2007;46:63–70.
[16] Ismail H, Osman H, Ariffin A. The effect of feldspar loading on curing characteristics, mechanical properties, swelling behavior and morphology of natural rubber vulcanizates. Int J Polym Mater 2005;54:43–62.
[17] Sukumar R, Menon ARR. Organomodified kaolin as reinforcing filler for natural rubber. J Appl Polym Sci 2004;107:3476–83.
[18] Escocio VM, Visconte Y, Munes RCR, De-Oliveira MG. Rheology and processability of natural rubber composites with mica. Int J Polym Mater 2008;57:374–82.
[19] Choi SS, Ha SH. Water swelling behaviors of silica-reinforced NBR composites in deionized water and salt solution. J Ind Eng Chem 2010;16:238–42.
[20] Da-Costa HM, Visconte IIR, Nunes RCR, Furtado CRG. Rice-husk-ash-filled natural rubber. II. Partial replacement of commercial fillers and the effect on the vulcanization process. J Appl Polym Sci 2003;87:1405–13.
[21] Murphy LJ, Wang MJ, Mahmud K. Carbon-silica dual phase filler: Part III: ESCA and FTIR characterization. Rubb Chem Technol 1998;71:998–1014.
[22] Zhang P, Wang MJ, Kutsovsky Y, Mahmud K. Carbon-silica dual phase filler application to tread compounds. Rubb World 2002;226:43–8.
[23] Rattanasom N, Saowapark T, Deeprasertkul C. Reinforcement of natural rubber with silica/carbon black hybrid filler. Polym Test 2007;26:369–77.
[24] Ismail H, Osman H, Jaafar M. Hybrid-filler filled PP/NR composites: effects of natural weathering on mechanical and thermal properties and morphology. J Vinyl Addit Technol 2008;14(1):142–51.
[25] Praveen S, Chattopadhyay PK, Albert P, Dalvi VG, Chakraborty BC, Chattopadhyay S. Synergistic effect of carbon black and nanoclay fillers in styrene butadiene rubber matrix: development of dual structure. Composites Part A 2009;40:309–16.
[26] Abdul-Kader M, Changwoon N. Influence of clay on the vulcanization kinetics of fluoroelastomer nanocomposites. Polymer 2004;45:2237–47.
[27] Jacob M, Thomas S, Varughese KT. Natural rubber composites reinforced with sisal/oil palm hybrid fibers: tensile and cure characteristics. J Appl Polym Sci 2004;93:2305–12.
[28] Leong YM, Bakar MBA, Ishak ZAM, Ariffin A. Characterization of talc/calcium carbonate filled polypropylene hybrid composites weathered in a natural environment. Polym Degrad Stab 2004;83:411–22.
[29] Ahmed K, Nizami SS, Raza NZ, Habib F. The effect of silica on the properties of marble sludge filled hybrid natural rubber composites. J King Saud Univ (Science) 2013;25:331–9.
[30] Ahmed K, Nizami SS, Raza NZ, Mahmood K, Kamaluddin S. An assessment of rice husk ash modified, marble sludge loaded natural rubber hybrid composites. J Mater Environ Sci 2013:205–16.
[31] Ahmed K, Nizami SS, Raza NZ. Characteristics of natural rubber hybrid composites based on marble sludge/carbon black and marble sludge/rice husk derived silica. J Ind Eng Chem 2013;19:1169–76.
[32] Ahmed K, Nizami SS, Raza NZ, Mahmood K. Effect of micro sized marble sludge on physical properties of natural rubber composites. Chem Ind Chem Eng Quart 2013;19:281–93.
[33] Malini KA, Kurian P, Ananthraman MR. Loading dependence similarities on the cure time and mechanical properties of rubber ferrite composites containing nickel zinc ferrite. Mater Lett 2003;57:3381–6.
[34] Govindjee S, Simo JC. A micro-mechanically based continuum damage model for carbon black filled rubbers incorporating Mollins effect. J Mech Phys Sol 1991;39:87–112.
[35] Wang MJ. Effect of polymer–filler and filler–filler interactions on dynamic properties of filled vulcanizates. Rubb Chem Technol 1998;71:520–89.
[36] Fan R, Zhang Y, Huang C, Zhang Y, Fan Y, Sun K. Effect of crosslink structures on dynamic mechanical properties of natural rubber vulcanizates under different aging conditions. J Appl Polym Sci 2001;81:710–8.
[37] Bhowmick AK, White JR. Thermal, UV- and sunlight ageing of thermoplastic elastomeric natural rubber–polyethylene blends. J Mater Sci 2002;37(23):5141–51.
[38] Ahagon A, Kida M, Kaidou H. Aging of tire parts during service. I. Types of aging in heavy-duty tires. Rubb Chem Technol 1990;63(5):683–97.
[39] Baldwin M, Bauer DR, Ellwood KR. Accelerated aging of tires, Part II. Rubb Chem Technol 2005;78(2):236–53.
[40] Clarke J, Ngolemasango EF, Bennett M. Kinetics of the effect of ageing on tensile properties of a natural rubber compound. J Appl Polym Sci 2006;102(4):3732–40.
[41] Osabohien E, Egboh SHO. Utilization of bowstring hemp fiber as a filler in natural rubber compounds. J Appl Polym Sci 2008;107(1):210–4.
[42] Blow CM, Hepburn C. Rubber technology and manufacture. 2nd ed. London: Butterworths; 1981.