Universiti Teknologi MARA
Cawangan Perlis
Jurnal Intelek

Volume 14 Issue 2 (December 2019)

Effect of Marble Tile Waste Powder (MTWP) Filler on Cure and Mechanical Behaviour of Natural Rubber (NR) Compound

Nor Mazlina Abdul Wahab[1*], Aztikah Azman[2], Noor Aishatun Majid[3], Sharifah Nafisah Syed Ismail[4], Mohd Syamaizar Mustafa[5]

[1,2,3,4,5] *Faculty of Applied Sciences, Universiti Teknologi MARA, 02600 Arau, Perlis, Malaysia*

*Authors' Email Address: *[1]normazlina@uitm.edu.my, [2]aztikah0208@gmail.com, [3]nooraishatun@uitm.edu.my, [4]syarifahnafisah@uitm.edu.my, [5]syamaizar@uitm.edu.my*

Received Date: 1 November 2019
Accepted Date: 30 November 2019

ABSTRACT

As part of the increasing effort to reutilize of residue generated from industrial activities, an attempt has been made to utilize marble tile waste as a potential filler in natural rubber (NR) composite when low strength applications are required. In the experimental part, 10 µm size of marble tile waste powder (MTWP), with five different loadings up to 40 parts per hundred of rubber (phr) were used in NR composites to study the effect of MTWP loading on the cure characteristics and mechanical properties. The addition of MTWP as filler resulted in shorter cure time and scorch time which is preferable in rubber processing. It also enhanced the properties of tensile modulus and hardness while tensile strength, elongation at break and swelling ratio decreased with increasing MTWP loading. This study thus shows that MTWP can be used as economical alternative filler in NR compound and has potential as a partial replacement of reinforcing filler like carbon black in various rubber products.

Keywords: *natural rubber, filler, composite, marble tile waste*

INTRODUCTION

In compounding rubber, filler is added to increase the mechanical and physical properties such as tensile strength, tear strength, hardness and abrasion resistance. Carbon black, silica, and other non-black fillers are commonly used fillers in rubber compounding. Carbon black has weak filler-filler interaction; thus, it will be easier to disperse in rubber, and this will cause increasing mechanical and physical properties of rubber vulcanizate (Ika et al., 2015). The performance of rubber can be modified by incorporation with different types of filler in consideration of various factors to develop recipe of rubber compound. One of the most important elements needs to be considered is cost. The cost must be relatively lower and at the same time, the productivity and processing time must not be affected. Therefore, recently, low cost filler which is easily obtained from agricultural, food industry and industrial waste like rice husk, eggshell, and marble tile waste have gain attention by many researchers in investigating their effects as potential replacement of conventional or synthetic filler in development of rubber compounded material. This step also tends to preserve the environment against uncontrolled pollution including water and air that give negative impact on our lives. The amount of industrial waste such as marble tiles waste should be minimized since it causes environmental problems such as water pollution and air pollution. Marble tiles waste could be found easily in the construction area. Marble waste is widely used in making roads and building construction material as the waste will form lightweight concrete, cut down the cost and increase

ISSN: 2231-7716
DOI: http://10.24191/ji.v14i2.238

the hardness of the concrete due to the coarse aggregates. Hence, the concept of recycle, reuse and reduce in waste management system is a good practice in order to reduce the impact of these materials to the environment due to urbanization.

Recent studies used marble waste in the form of marble sludge and marble dust to reinforce polymer matrix. Marras and Careddu (2018) have investigated the effect of dewatered marble sludge in tyre compound as compared with commercial calcium carbonate filler with the objective to use waste of marble sludge as filler in producing high added value product. Awad and Abdellatif (2019) and Awad et al., (2019) reinforced thermoplastics materials i.e. low density polyethylene (LDPE) and polypropylene (PP), respectively, with marble dust in producing novel composite material. Positive results obtained shows LDPE reinforced with marble dust improved mechanical and thermal properties of the composite. Similar trend observed in PP matrix has also shown significant influence of this waste material. In addition, in 2017 Borazan and Gokdai used waste marble dust as reinforced material in polyester based hybrid composite and found that the blend of waste marble dust with feather fiber has significant effect on mechanical properties of composites.

Marble tiles waste powder (MTWP) is rarely used in rubber compounding. Hence, an effort has been made in this study to use MTWP as a potential filler in natural rubber (NR) compound and its’ effect on curing, mechanical and swelling behavior of rubber vulcanizate has been studied.

METHODOLOGY

In this project, the main raw materials used are natural rubber (SMR10) and fixed amount of carbon black (CB) filler along with different amounts of marble tiles waste powder (MTWP). Other compounding ingredients used are zinc oxide (ZnO) as an activator, stearic acid as co-activator, mercaptobenzothiazole (MBT) as accelerator, 2,2,4-trimethyl-1,2-dihydroquinoline (TMQ) as an antioxidant and sulphur as vulcanizing agent. All these materials were supplied by Zarm Scientific & Supplies Sdn. Bhd., Butterworth, Pulau Pinang, except for MTWP that was prepared in the laboratory at UiTM Perlis Branch. MTWP was obtained from industrial area at Perak and prepared in UiTM laboratory.

Formulation

The formulations shown in Table 1 were used in this study.

Table 1: Recipe Used to Produce NR Filled MTWP compounds

Materials	Composition (phr)
Natural Rubber (NR)	100.0
ZnO	5.0
Stearic Acid	2.0
MBT	2.4
TMQ	1.5
Sulphur	1.6
CB	40.0
MTWP	0, 10, 20, 30, 40

The amount of MTWP was varied. The amount used was increased in each SMR/MTWP compound formulations (0, 10, 20, 30, 40 phr). All SMR compounds were cured by a semi-efficient curing system (semi-EV) to get a compromise in product performance and cost.

Preparation of Marble Tile Waste Powder (MTWP)

Marble tiles waste was collected around the construction area in Perak, Malaysia and they were cleaned from sand and cement. Then, it was cut into small pieces to make the grinding process easier. The marble tile waste was oven dried for 24 hours at 150ºC. Then, it was grounded into powder form in a hammer mill. The powder was then sieved with 10 μm size sieve.

Preparation of MTWP Filled NR Compounds

Two-roll mill was used to soften the SMR and preparation of rubber compound based on the formulations in Table 1. From the control formulation, an increased amount of MTWP was added into the compound. All samples were milled at room temperature. Once SMR softened, other ingredients then were added until homogeneous compound visually observed, within 30 min. The accelerator and sulphur were added at the end of the mixing process to avoid premature crosslinking reaction occur. After 24 hours, the prepared compound underwent cure tests to obtain its optimum cure time before further proceeded with molding process and formed into sheet of 1 mm thickness via hot press at 170ºC. Rheometer-Hung Ta HT-M2000 was operated at 170ºC, 20 min. for cure characteristics (cure time and scorch time) determination, according to ASTM D2084-79.

Tensile Properties

Tensile test was carried out according to ASTM D412 using a Universal Testing Machine from Instron Co., USA. Sample with 1 mm thickness dumb-bell shape specimens, 3 mm width were cut by using dumb-bell cutter/die. The gauge length was fixed at 25 mm and the test was performed at laboratory temperature, 25±3 ºC with a crosshead speed of 500 mm/min. Tensile properties of five identical samples for each formulation were measured and the average values of tensile strength, tensile modulus, and elongation at break were reported, accordingly.

Hardness Test

Hardness test was conducted using Durometer Hardness Tester (Shore A) according to ISO 868. Each formulation consists of two rubber samples and each sample was tested at five different points to determine its average value.

Swelling Test

The swelling test was conducted by soaking about 0.2-0.25 g of rubber sample into pure toluene in accordance with ISO1817. The rubber test samples were weighed via weighing balance and allowed to swell until reach diffusion equilibrium, which took 120 hours (5 days), at room temperature. After that, samples were dried until constant weight was obtained in the oven at 60 ºC. The swollen weight was measured, and the result was recorded every day until day five. The swelling coefficient of the sample was recorded and calculated. Swelling index (%) calculated using the formula below:

Swelling index (%): $(W_1 – W_2) / (W_2) \times 100$
where,
W_1: Swollen weight
W_2: Initial weight

RESULTS AND DISCUSSION

Effect of MTWP Loading on Cure Characteristics of NR Compound

Figure 1 and 2 show the effect of different loading of MTWP on cure time (t_{c90}) and scorch time (ts_2) of NR compound. As the MTWP loading increased, the t_{c90} and ts_2 gradually decreased. The heat generated from the mixing process and high shear stress causes the breaking layer of fillers. According to Mohd et al., (2016), heat is generated from shear force during mixing and does caused the scorch time to decrease. There is development of heat and energy, and filler interactions occur during mixing process. The development of heat and energy produced increases as the amount of MTWP increases during compounding process. Therefore, as the amount of MTWP increases, the time taken for compounding process increases, thus, heat and energy produced are also increased. According to the Ahmed (2015), during compounding, high energy and heat produced, the viscosity increases with the better shear heating of the rubber compound and this causes the decreased of t_{c90} and ts_2 of the compound. In addition, according to Ahmed et al., (2013) the interaction of MTWP and dispersion of filler cause the vulcanization to become more effective and resulting in decreasing of scorch time and cure time of the NR compound. The lowest t_{c90} and ts_2 at 40 phr MTWP indicates that there is the highest development of heat and energy and effective interaction occurs between the MTWP with NR at this composition.

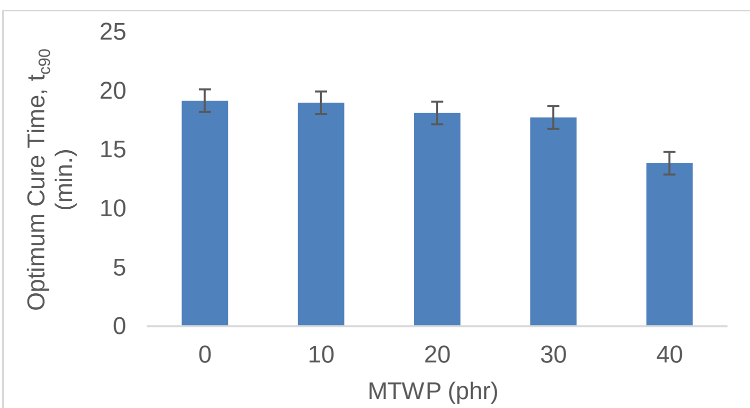

Figure 1: Optimum Cure Time, t_{c90} (min.) of MTWP Filled NR Compound

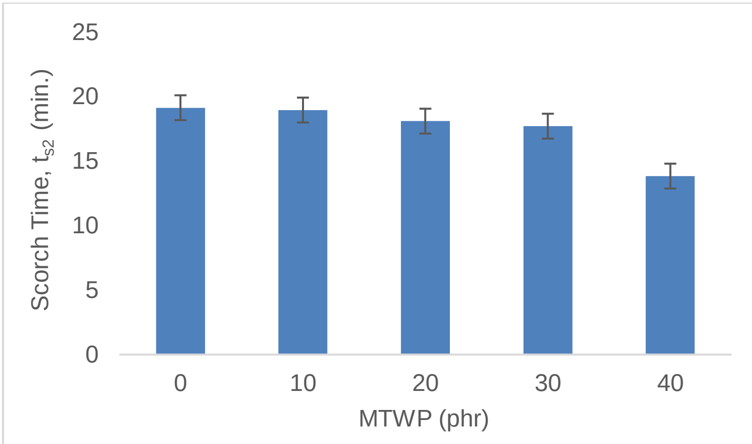

Figure 2: Scorch Time, ts_2 (min.) of MTWP Filled NR Compound

Effect of MTWP Loading on Mechanical Properties of NR Compound

Figure 3 reveals the trend of tensile strength for MTWP filled NR. The increase of MTWP loadings leads to gradually decreased on tensile strength of NR compound than that of the controlled compound. This may be caused by the agglomeration of MTWP particles when increased at higher loadings. According to Ahmed (2015), agglomeration of filler particles in rubber compound may cause by the large particle size. In addition, Ika et al., *(*2015) states that fillers with particle size greater than 10 µm must be avoided because it will drop the performance of rubber compound. Meanwhile, Ahmed et al., (2014) claims that the size of the particle which is usually large, have a possibility to interrupt matrix continuity, thus, cause the decrement of effective load bearing across the section area. In addition, for maximum reinforcement, the particles of fillers must have the same size or smaller than the chain end to end of matrix distance, so that the degree of reinforcement will increase with decrease of particle size or increase in surface area of filler. Applied stress is transferred from the polymer matrix to the strong and stiff mineral. Stress transfer will be better affected if the mineral particle is smaller and have greater surface area so that the possibility of interruption toward matrix continuity reduced (Ngamsurat et al., 2011).

In addition, decreased tensile strength is probably due to weak filler-rubber interaction. It happens due to the dilution effect and marble sludge (MS) filler agglomeration. The filler-rubber interaction increases till the optimum loading, but further filler addition causes a stronger filler-filler interaction as compared to the filler-rubber interaction (Ahmed et al., 2012a). Hence, in this study, the decreased in tensile strength has indicated that the filler-rubber interaction becomes weak as increased amount of MTWP in NR compounds.

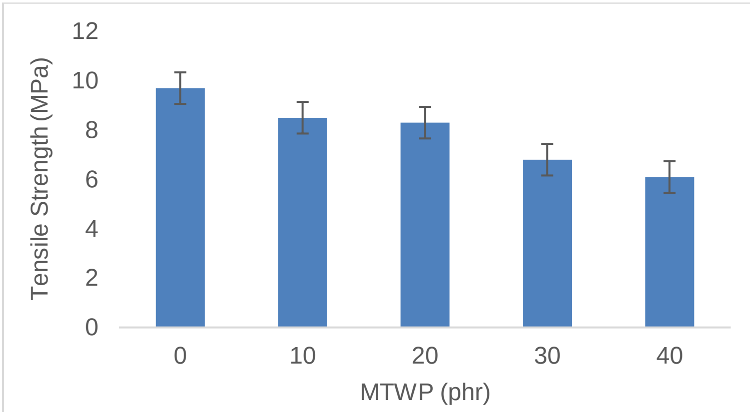

Figure 3: Tensile Strength (MPa) of MTWP Filled NR Compound

The tensile modulus of MTWP filled NR as shown in Figure 4 gradually increased as the filler loading increased from 10 to 40 phr. It shows that the incorporation of MTWP resulted on an increase in stiffness of NR compound. There is reduced amount of rubber matrix as increased amount of MTWP in NR compound. Hence, it could be said that the elasticity of the compound also tends to decrease, where it becomes more stiff and harder as the amount of MTWP increased. The increased moduli of the filled NR composites are due to the reduced elasticity and increased rigidity of the rubber matrix (Ahmed et al., 2012a). In addition, the increasing of tensile modulus of NR compound also cause by better dispersion of carbon black (CB) and marble sludge waste powder in the rubber matrix (Ahmed, 2015). Hence, the stiffness of the NR compound increases as the dispersion between rubber and filler increases.

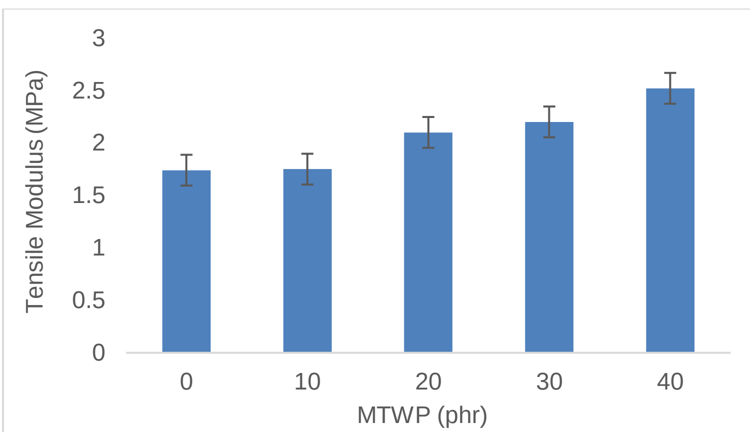

Figure 4: Tensile Modulus (MPa) of MTWP Filled NR Compound

It is clearly seen that the elongation at break, E_b (%) of NR compound in Figure 5 decreased as MTWP loading increased. This is attributed to the decreasing of ductility of the compound as increase amount of MTWP. Based on Ahmed et al., (2013), the reduction on % E_b of marble waste powder hybrid with CB indicates that the ductility becomes less and the presence of conventional filler such as CB causes the restriction in molecular chains movement. Molecular chain movement is limited because of the interaction between the filler and the natural rubber molecules that lead to the development of crosslinking structure. Thus, free movement of molecules become limited and there is an increase in the resistance to shape changes when tension is applied. In addition, Ahmed et al., (2012b) claimed that the reduction in % E_b of Silica/Marble sludge in NR composite caused by the high resistance against stretching when strain is applied. High resistance causes by the adherence of the filler to the polymer that leads to the increased stiffness of compound. This result, therefore, agrees with the obtained tensile modulus result for MTWP filled NR compound in this study.

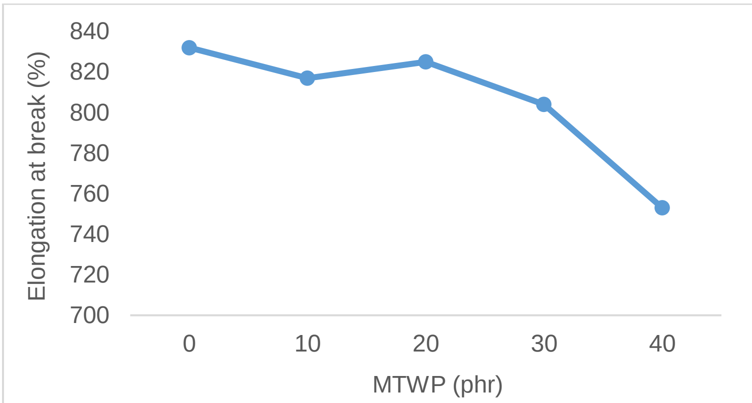

Figure 5: Elongation at Break (%) of MTWP Filled NR Compound

Effect of MTWP Loading on Hardness (Shore A) of NR Compound

Figure 6 demonstrates the results for hardness (Shore A) of MTWP filled NR compound. It is clearly seen that there are gradually increments in hardness of NR compound as MTWP loading increased from 10 to 40 phr. The hardness was influenced by the stiffness of the compound. Since there are increased in stiffness of rubber compound, thus the rigidity of the rubber compound also increased when the elasticity of rubber reduced. Therefore, it finally tends to result in higher hardness value of the NR compound. This result agrees with Mohd (2016). He states that the hardness of the rubber compound that consists of a combination of marble sludge waste/rice husk increased as the ratio of marble sludge increased in the compound and the hardness of rubber is influence by the better rigidity of rubber compound. This is because the agglomeration of marble sludge waste occurs and causes the increasing of mass ratio. This result is parallel with increasing of modulus observed for every compound in this study. Increase in modulus means that there are increase in stiffness, which finally increase in hardness of the compound as well. According to Brown and Soulagnet (2001), hardness is essentially a measure of modulus. On top of that, Ahmed et al., (2012a) finds that the hardness of NR composites increases with the increase in MS content, which is resulted from more rigid compounds.

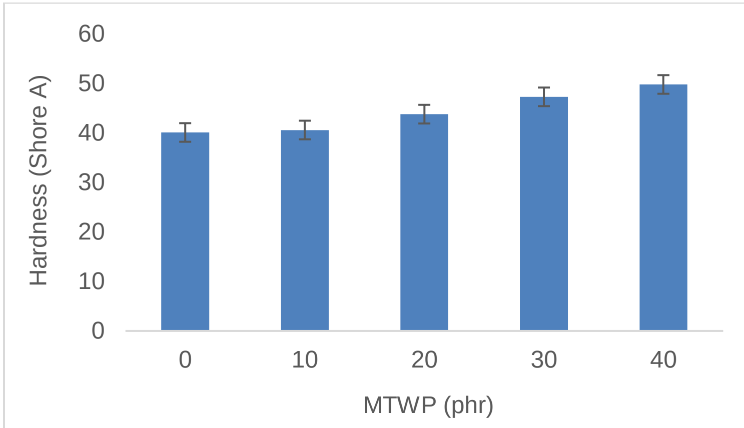

Figure 6: Hardness (Shore A) of MTWP Filled NR Compound

Effect of MTWP Loading on Swelling Index (%) of NR Compound

The influence of MTWP loading on the swelling index (%) of NR compound is shown in Figure 7. The result shows the relationship of the swelling index (%) of toluene uptake for five days for each of NR compounds. Generally, the result shows that the swelling index decreases as the amount of MTWP increases. This is because the increasing amount of MTWP in rubber matrix resulted in decreased amount of matrix constituent which tend to reduce flexibility or reduction of molecular movement of the rubber. Hence, the penetration of toluene into rubber matrix will be difficult. As according to Ahmed et al., (2012a) the penetration of toluene into MS-filled NR composites are reduced with the increment of Marble Sludge (MS) loading. This means that higher amount of MS loading restricted the penetration of toluene in filled NR composites. The similar trend also reported by Ahmed et al., (2012b), whose states that the penetration of toluene into MS/NR/Silica hybrid composites is reduced by the increasing silica content. This means that higher amount of silica restricted the penetration of toluene in composites.

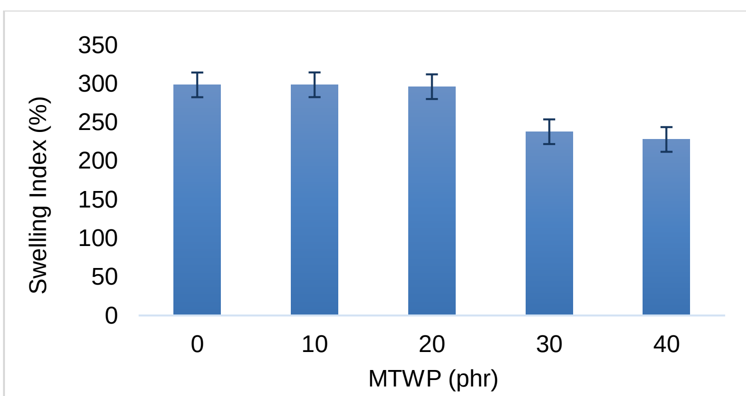

Figure 7: Swelling Index (%) of MTWP Filled NR Compound

CONCLUSION

In conclusion, the influence of MTWP loading as filler in NR compound on cure characteristics, mechanical properties, and swelling ratio had been investigated. Optimum cure time, scorch time, tensile strength, elongation at break and swelling ratio of the compound is reduced, meanwhile tensile modulus and hardness are increased by increasing the amount of MTWP in NR compound. The NR compound becomes stiffer as MTWP loading increased, indicate an increased in rigidity as decreased in ductility. In addition, less flexibility of NR compound obtained which shows by increased modulus and hardness of the compound. Hence, MTWP has good potential usage as filler in NR compound, especially for low strength applications.

REFERENCES

Ahmed, K., Sirajuddin, N. S., Zahid, R. N., & Mahmood, K. (2012a). Mechanical, swelling, and thermal aging properties of marble sludge-natural rubber composites. International Journal of Industrial Chemistry, 3, 21. http://www.industchem.com/content/3/1/21

Ahmed, K., Sirajuddin, N. S., Zahid, R. N., & Habib, F. (2012b). The effect of silica on the properties of marble sludge filled hybrid natural rubber composites. Journal of King Saud University, 25, 331-339. https://doi.org/10.1016/j.jksus.2013.02.004

Ahmed, K., Sirajuddin, N. S., Zahid, R. N., Kamaluddin, S., & Mahmood, K. (2013). An assessment of rice husk ash modified, marble sludge loaded natural rubber hybrid composites. Journal of Material Environment Science, 4(2), 205-216.

Ahmed, K., Sirajuddin, N. S., & Zahid, R. N. (2014). Reinforcement of natural rubber hybrid composites based on marble sludge/silica and marble sludge/rice husk derived silica. Journal of Advanced Research, 5, 165-173. https://doi.org/10.1016/j.jare.2013.01.008

Ahmed, K. (2015). Hybrid composites prepared from industrial waste: Mechanical and swelling behaviour. Journal of Advance Research, 6, 225-232. https://doi.org/10.1016/j.jare.2013.12.002

Borazan, A. A. & Gokdai, D. (2017). Polymer composites reinforced with waste marble dust and fibers from chicken feathers as an alternative material. Research Gate Publication: Fresenius Environmental Bulletin, 26, 2095-2103.

Awad, A. H. & Abdellatif, M. H. (2019). Assessment of mechanical and physical properties of LDPE reinforced with marble dust. Journal of Composites Part B: Engineering, 73, 106948. https://doi.org/10.1016/j.compositesb.2019.106948

Awad, A. H., Ramadan El-Gamasy, Ayman, A. Abd El-Wahab, & Abdellatif, M., H. (2019). Mechanical behavior of PP reinforced with marble dust. Construction and Building Materials, 228, 116766. https://doi.org/10.1016/j.conbuildmat.2019.116766

Brown, R. P., & Soulagnet, G. (2001). Microhardness profiles on aged rubber compounds. Polymer Testing, 20, 295-303. doi:10.1016/S0142-9418(00)00035-0

Marras, G. & Careddu, N. (2018). Sustainable reuse of marble sludge in tyre mixtures. Resources Policy, 59, 77-84. https://doi.org/10.1016/j.resourpol.2017.11.009

Ika, M. U., Riastuti, F., Rahayu, S., Fitriani, D. A., & Saputra, D. A. (2015). Influence of carbon black and silica filler on the rheological and mechanical properties of natural rubber compound. Journal of Procedia Chemistry, 16, 258-264. https://doi.org/10.1016/j.proche.2015.12.053

Mohd (2016). Effect of filler loading on curing characteristics and tensile properties of palygorskite natural rubber nanocomposite. Procedia Chemistry, 9, 351-358. https://doi.org/10.1016/j.proche.2016.03.023

Ngamsurat, S., Boonkerd, K., Leela-adisorn, U., & Potiyaraj, P. (2011). Curing characteristics of natural rubber filled with gypsum. Journal of Energy Procedia, 9, 452-458. https://doi.org/10.1016/j.egypro.2011.09.051