

Effects of Coconut Coir Powders on the properties of Natural Rubber Composites

Teerakorn Kongkaew¹, Sureeporn Kumneadklang¹,

Jate Panichpakdee¹ and Siriporn Larпкиattaworn¹ *

Received May 18, 2020; Revised, June 10, 2020; Accepted June 29 2020

Abstract

In this work, the coconut coir powders (CCP)/natural rubber (NR) composites were successfully prepared. The CCP acts as a reinforcing filler with two different sizes of fine powders (39 μm) and coarse powders (101 μm). The coconut coir powders added to the natural rubber matrix at the filler content of 25, 50, 75, and 100 phr. The effect of CCP contents on physical and mechanical properties was studied. The result revealed that the increase in CCP content has decreased the tensile strength, elongation at break and toughness of composites but increased the modulus of elasticity. The CCP/NR composite at 25 phr of filler loading shows better mechanical properties. For different sizes of fillers, the F-CCP exhibit the better mechanical properties and hardness compare to C-CCP. These properties of composites indicate that it can develop and possible apply in rubber mats.

Keywords: Coconut coir powders, Natural rubber, Mechanical properties, Composites

Introduction

Polymer composites are a combination of a polymer matrix with fillers. They have been attractively applied in industrial and academic researches due to the control of the material properties. For the past few decades, researchers have shown increasing interest in composite materials for those of biodegradable, eco-friendly, and renewable. Natural rubber (NR) as a green and renewable polymer is used extensively in many applications, including in tires, automotive parts, and rubber floor mats, because of its excellent mechanical and elastic properties. However, fillers are essential to modify the NR properties for the versatile application. The capability of the reinforcing filler depends on various factors such as surface area, the shape of fillers, and particle size. Natural fillers have been in enormous demand as a reinforcing material. These fillers act as good reinforcing agents with several specific properties such as high toughness, low cost, lightweight, good specific strength properties, and modulus, and complete burning without residue on the combustion (Ismail et

al., 2002; Luz et al., 2007; Panthapulakkal et al., 2006). The natural fibers act as reinforcing natural fillers with biodegradable and renewable properties. The probability of natural fibers such as kenaf, sisal, pineapple leaf, banana jute to produce polymer composites was studied (Faruk et al., 2012). Coconut coir is an inexpensive agricultural product that can be obtained in a large volume from the local community. It is a cheap eco-friendly fiber that is also less expensive than jute and sisal (Geethamma et al., 1998). The utilization of coconut coir as reinforcing fillers in polymer composites becomes more desirable due to their high strength and modulus properties (Macedo et al., 2010; Monteiro et al., 2008). Studies on powdered coconut coir as the filler has been appeared in the literature (Kaewduang et al., 2015; Sarki et al., 2011). This work aims to investigate the possibility of Coconut coir powders (CCP) of different particle sizes in using as reinforcement filler in the NR matrix. The effect of filler content on the properties of CCP/NR composites was investigated.

¹ Thailand Institute of Scientific and Technological Research, KhongLuang, Pathum Thani, 12120, Thailand

* Corresponding author: Email:siriporn@tistr.or.th

Materials and Methods

Preparation of Coconut coir powders

The coconut coir was collected from the shell of the matured coconut and was broken into small pieces. The coconut coir was oven-dried at 100 °C overnight for removing moisture. After drying, a portion of the coconut coir powders (CCP) was milled into fine particles by the blender machine and fed into a vibrating sieving machine using 300 μm mesh which was called Fine CCP (F-CCP). A second portion, CCP were sieved through the previous mesh size without milling which was called Coarse-CCP (C-CCP). Their powder sizes were measured by an optical microscope and were used as the filler in NR composites. The average powder sizes of F-CCP and C-CCP were about 39 μm and 101 μm , respectively. And the morphology is shown in Figure 1.

Preparation of the composites

The formulation of the mixes was given in Table 1. The natural rubber used for the study was crepe rubber grade obtained from the local rubber companies. Coconut coir powders (CCP) were used as the filler. The composite compounds with different CCP loading were mixed using an internal mixer. The Sulphur and TMTD acted as a vulcanizing agent and were added during the milling step using a two-roll milling machine. The sheeted rubber compounds were kept for maturation at room temperature for 24 h until the mixture was hardened. After that, the rubber composite compounds were vulcanized in the compression-molding under 1

min preheating at 150 °C and compressed under a pressure of 75 kg/cm^2 (mold dimension: 160 x 160 x 2 mm^3).

Characterization

Mechanical properties

For the stress-strain behavior of composite samples, the Shimadzu Testing Machine (AG-X Plus 10kN model) was used. The tensile properties of the composites were tested according to ASTM D412-1998, at a crosshead speed of 500 mm/min at room temperature. The Hardness test on Shore-A Durometer according to ASTM D2240 was conducted using Bareiss Digi test II hardness tester. The specimen should be at least 6 mm thick and hardness reading was performed within three seconds during testing. Finally, the hardness was measured for five different positions and the average values were presented.

Physical properties

The swelling measurements of the composites were examined in the toluene. The specimen with a thickness of 2 mm was cut into a rectangular shape in the dimension of 25x25 mm. Then the composite samples were dried at 60°C for 24 h. The specimen was immersed in toluene for 48 h at 25°C. The swollen composite sample was taken out of the toluene and wiped with tissue paper to remove excess toluene. Then, the weight of the swollen samples was determined precisely. This data can be determined for the percentage of

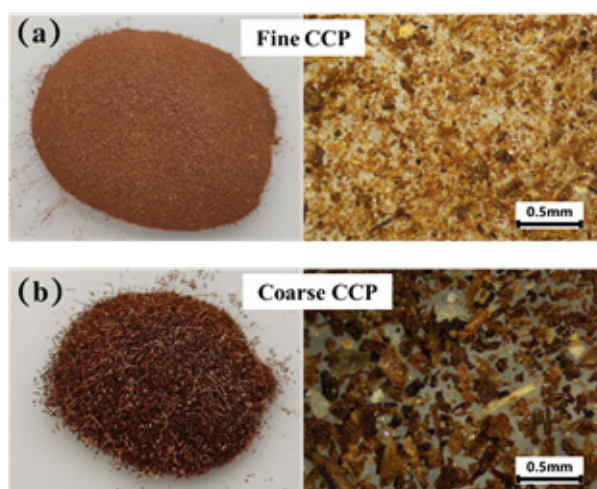


Figure 1. (a) CCP with milling by blender machine (F-CCP) and (b) CCP without milling (C-CCP) and both sieving through 300 μm sieve size

Table 1. Mixing formulations of composites

Ingredient	phra
Crepe rubber (NR)	100
ZnO	5
Stearic acid	2
Wingstay L	1
Ozone wax	1
Clay	100
White oil	1
CCPb	0, 25, 50, 75, 100
TMTDc	1
Sulfur	1.7

a Parts per hundred of rubber

b Coconut CoirPowders of both fine and coarse in particle sizes

c Tetramethylthiuram Disulfide

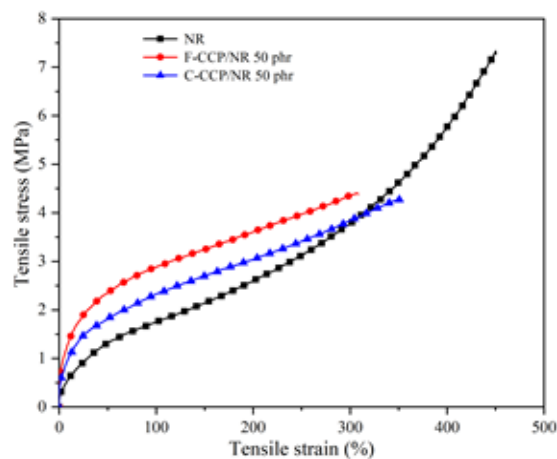


Figure 2. Stress-strain curves of NR, F-CCP/NR and C-CCP/NR composites with CCP content of 50 phr.

swelling and also rubber-filler interactions. The percentage of swelling can be calculated by;

$$\text{Swelling (\%)} = \frac{M - M_d}{M_d} \times 100 \quad (2)$$

Where M and M_d are the wet weight after removing the excess toluene and dry weight, respectively.

Results and discussion

Figure 2 shows the tensile stress-strain curves in CCP/NR composites with 50 phr of F-CCP and C-CCP. The NR sample without CCP loading exhibits a typical

stress-strain behavior of vulcanized rubber showing the ultimate strain of around 440%. It is noted that all the CCP/NR composites have higher moduli than the NR sample. In the presence of 50 phr of C-CCP/NR composite, the ultimate strength and strain are reduced. Tensile strength was decreased by about 42% and strain at the break by 23%.

The CCP with the massive diameter causes deterioration in the properties due to the formation of voids at the CCP-matrix interface because of insufficient interfacial interaction (Parambath et al., 2019). Besides, the F-CCP/NR composite was observed that the tensile

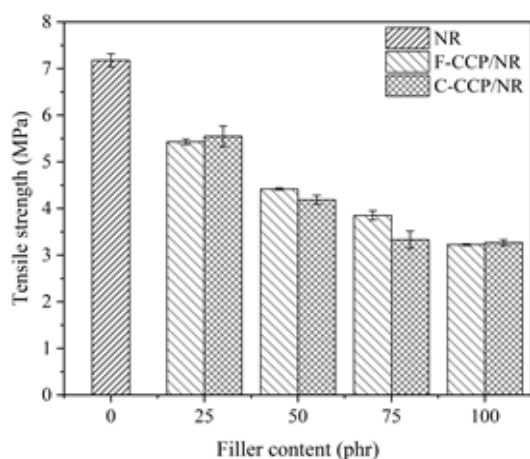


Figure 3. The effect of filler content on tensile strength of NR, F-CCP/NR and C-CCP/NR composites

strength can be improved. Although the value of elongation at break decreased from 341% to 311%, however, the F-CCP/NR composites retain the elastic properties of the pure NR

The effect of filler loading with different CCP sizes on tensile strength of CCP/NR composites is shown in Figure 3. As observed, the increase of CCP contents affected in the weakened tensile strength when compared with the NR compound. For the CCP/NR composites, the increase of CCP content inducing tensile strength decreased. It is found that the maximum value of tensile strength at 25 phr of filler content. This result implies that the CCP/NR composite at 25 phr uniform the dispersion of filler in NR compounds and might be the effect of better interfacial adhesion due to a large interfacial area of contact contributes to a higher tensile strength (Balan et al., 2017; Herrera-Franco et al., 2004). When the filler contents are applied above 25 phr, the tensile strength decreased due to weak interaction and bonding between the filler and NR matrix are responsible for the decline of tensile strength. The other reason might be due to agglomeration. And, therefore, the filler – filler interaction of the CCP filler also increases (Balan et al., 2017). This is consistent with the study of Leha et al. which presents the effect of filler from 10 to 40 %. The filler content at 25% showed the highest tensile strength. For more addition of the filler, it presented a decrease in mechanical properties (Noor Leha et al., 2014). For the comparison of the tensile strength between F-CCP/NR and C-CCP/NR composites, it was

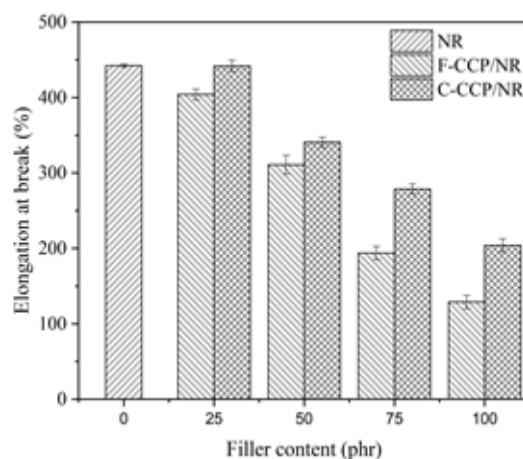


Figure 4. The effect of filler content on elongation at break of NR, F-CCP/NR and C-CCP/NR composites.

found that the filler content at 25 and 100 phr, C-CCP/NR exhibited the value which closes to the value of F-CCP. However, the filler contents of 50 and 75 phr exhibit the tensile strength of F-CCP/NR higher value than C-CCP/NR. This is due to the effect of the high specific surface area of filler sizes on the NR matrix (Sareena et al., 2012).

Figure 4 shows the elongation at break of the CCP/NR composites is decreased with increasing filler content due to the addition of filler which reduces the mobility and increases the brittleness of the composites (Islam et al., 2017).

The lower elongation at high filler content might be due to the void portion is filled up according to the adding of filler content. Besides, the increase of CCP filler content in the NR compounds stiffens and also hardens the NR compounds. It will reduce resilience and toughness that leads to lower elongation. The toughness behavior is showed in Figure5. Comparing the elongation at break at the different size of CCP loading, it is observed that F-CCP/NR composites provide the higher elongation than those of C-CCP/NR composites. This result implies that it is due to better interaction between F-CCP and NR matrix (Parambath et al., 2019; Wongsorat et al., 2014).

Figure 6. shows the effect of filler content on the modulus of elasticity of F-CCP/NR and C-CCP/NR composites. Fillers are known to enhance the modulus, exhibited that the modulus of the filler is higher than that of the NR matrix (0 phr) (Sareena et al., 2012). In

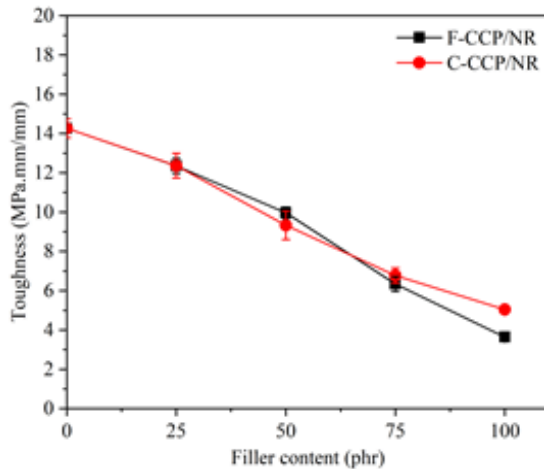


Figure 5. Effect of filler content on the toughness of F-CCP/NR and C-CCP/NR composites.

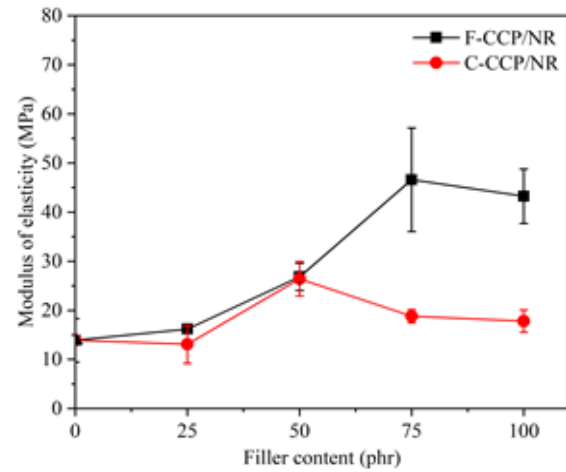


Figure 6. Effect of filler content on Modulus of elasticity of F-CCP/NR and C-CCP/NR composites.

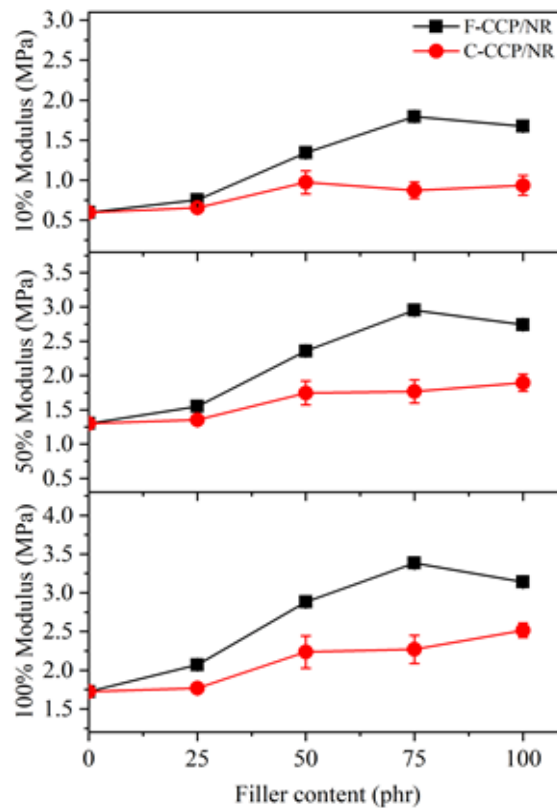


Figure 7. Moduli at (a) 10% strain, (b) 50% strain and (c) 100% strain of F-CCP and C-CCP filled NR composites

the case of F-CCP/NR, composites increase in the modulus with the filler content increasing almost linear up to 75 phr. While the modulus decreased when further increasing the filler content at 100 phr. For the C-CCP/NR composites, it exhibits an increase of modulus with the filler content up to 50 phr. Then an increasing filler content causes the modulus trend to decrease which is lower as compared with F-CCP/NR composites.

This result implies that modulus of F-CCP/NR composites improved due to the homogeneous distribution

and their effective interaction with the matrix restrain the molecular movement (Parambath et al., 2019). The agglomeration of filler at a higher loading does not act a disturbing effect on the modulus due to it is measured at low strains which the stress concentration is not enough for initiation of crack.

The effect of filler content on the reinforcing efficiency of F-CCP and C-CCP, moduli at 10%, 50%, and 100% strains of the composites are observed in Figure 6. This variation is a measure of the stiffness of compos-

ites. It can be shown that 10%, 50%, and 100% strains increase slightly with increasing filler content. This reveals that the filler powder acts like rigid particulates since it has a higher modulus than the NR matrix (Sareena et al., 2012). The moduli at 10%, 50% and 100% strains of the composites containing F-CCP were higher than those containing C-CCP. This indicates that the smaller sized filler in F-CCP/NR composites performed the higher modulus values than those of larger sized filler in C-CCP/NR composites.

Figure 8. shows the hardness of CCP/NR composites with different sizes and filler loading of CCP. Hardness increases with the increase in filler content. The hardness of F-CCP/NR composites exhibits higher values than C-CCP filled composites. Hardness is a measure of the resistance to deformation. The incorporation of fillers into an NR matrix reduces the elasticity of rubber. All fillers used are non-deformable solids and the addition of more rigid particles leads to increased rigidity and stiffness of the material.

The CCP/NR composites with different sizes of filler content characterized to examine swelling in toluene at room temperature for 48 h are presented in Figure 9. The highest swelling value is NR composite at 197% while the tendency swelling decreases with increasing of the filler loading in the NR composites. This implies that the filler loading in NR blends that restrict the molecular movement of the rubber.

This then made it more difficult for the toluene to penetrate through the rubber, thus, decreasing the swelling percentage. However, the swelling percentage decreased up to 50 phr in solvent uptake (%) at equilibrium swelling of CCP filled NR can be additionally described in the increase in contact surface area between polymer and filler.

At lower loading, the dominating effect is polymer-filler networks, whereas, at higher loading, the filler-filler networks will govern in the composites (Swapna et al., 2016). The result was related to the increase in tensile modulus, hardness, and also tensile strength for CCP filled NR composite.

Conclusion

In summary, this report presented the effect of particle sizes of fine and coarse coconut coir powders on the mechanical properties of NR and CCP/NR composites. All CCP/NR composites were prepared with 25, 50, 75, and 100 phr of CCP filler content. The increase in CCP content has decreased the tensile strength, elongation at break and toughness, whereas increasing the modulus of elasticity of CCP/NR composites. The result reveals that the CCP/NR composite of 25 phr filler loading exhibits better mechanical properties. F-CCP filler exhibits better mechanical properties and hardness as compared to C-CCP filler. The swelling values of CCP/NR composites in the toluene solution trend to decrease

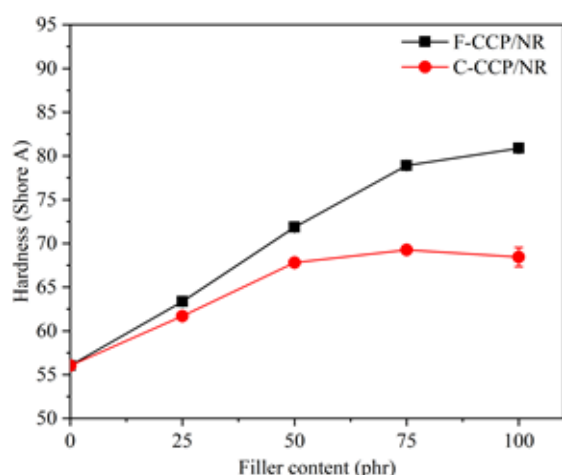


Figure 8. Variation of hardness with filler content for CCP/NR composites

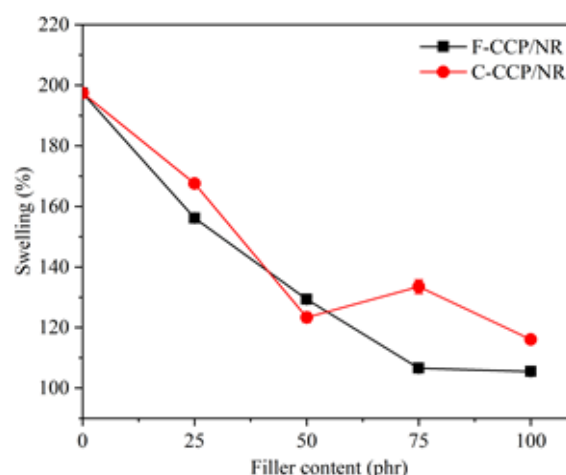


Figure 9. The effect of CCP loading on the swelling percentage of NR composites

with increasing filler loading in the NR composites. These results indicate that CCP/NR composites can be developed and apply for rubber mat.

Acknowledgments

The authors are grateful to The Expert Centre of Innovative Materials, Thailand Institute of Scientific and Technological Research (TISTR).

References

- Balan, A. K., Mottakkunnu Parambil, S., Vakyath, S., Thulissery Velayudhan, J., Naduparambath, S., & Etathil, P. (2017). Coconut shell powder reinforced thermoplastic polyurethane/natural rubber blend-composites: effect of silane coupling agents on the mechanical and thermal properties of the composites. *Journal of Materials Science*, 52(11), 6712-6725. doi:10.1007/s10853-017-0907-y
- Faruk, O., Bledzki, A. K., Fink, H.-P., & Sain, M. (2012). Biocomposites reinforced with natural fibers: 2000–2010. *Progress in Polymer Science*, 37(11), 1552-1596. doi:https://doi.org/10.1016/j.progpolymsci.2012.04.003
- Geethamma, V. G., Thomas Mathew, K., Lakshminarayanan, R., & Thomas, S. (1998). Composite of short coir fibres and natural rubber: effect of chemical modification, loading and orientation of fibre. *Polymer*, 39(6), 1483-1491. doi:https://doi.org/10.1016/S0032-3861(97)00422-9
- Herrera-Franco, P. J., & Valadez-González, A. (2004). Mechanical properties of continuous natural fibre-reinforced polymer composites. *Composites Part A: Applied Science and Manufacturing*, 35(3), 339-345. doi:https://doi.org/10.1016/j.compositesa.2003.09.012
- Islam, M., Das, S., Saha, J., Paul, D., Islam, M., Rahman, M., & Khan, M. (2017). Effect of Coconut Shell Powder as Filler on the Mechanical Properties of Coir-polyester Composites. *Chemical and Materials Engineering*, 5, 75-82. doi:10.13189/cme.2017.050401
- Ismail, H., Edyham, M. R., & Wirjosentono, B. (2002). Bamboo fibre filled natural rubber composites: the effects of filler loading and bonding agent. *Polymer Testing*, 21(2), 139-144. doi:https://doi.org/10.1016/S0142-9418(01)00060-5
- Kaewduang, M., Chaichana, E., Jongsomjit, B., & Jaturapiree, A. (2015). Use of coir-filled LLDPE as a reinforcement for natural rubber composite. Paper presented at the Key Engineering Materials.
- Luz, S. M., Gonçalves, A. R., & Del'Arco, A. P. (2007). Mechanical behavior and microstructural analysis of sugarcane bagasse fibers reinforced polypropylene composites. *Composites Part A: Applied Science and Manufacturing*, 38(6), 1455-1461. doi:https://doi.org/10.1016/j.compositesa.2007.01.014
- Macedo, J. d. S., Costa, M. F., Tavares, M. I. B., & Thiré, R. M. S. M. (2010). Preparation and characterization of composites based on polyhydroxybutyrate and waste powder from coconut fibers processing. *Polymer Engineering & Science*, 50(7), 1466-1475. doi:10.1002/pen.21669
- Monteiro, S. N., Terrones, L. A. H., & D'Almeida, J. R. M. (2008). Mechanical performance of coir fiber/polyester composites. *Polymer Testing*, 27(5), 591-595. doi:10.1016/j.polymertesting.2008.03.003
- Noor Leha, A. R., & Nordin, N. A. (2014). Effect of Filler Compositions on the Mechanical Properties of Bamboo Filled Polyester Composite. *Advanced Materials Research*, 879, 90-95. doi:10.4028/www.scientific.net/AMR.879.90
- Panthapulakkal, S., Zereshkian, A., & Sain, M. (2006). Preparation and characterization of wheat straw fibers for reinforcing application in injection molded thermoplastic composites. *Bioresource Technology*, 97(2), 265-272. doi:https://doi.org/10.1016/j.biortech.2005.02.043
- Parambath Kanoth, B., Thomas, T., Joseph, J. M., & Narayanankutty, S. K. (2019). Restructuring of coir to microfibers for enhanced reinforcement in natural rubber. *Polymer Composites*, 40(1), 414-423. doi:10.1002/pc.24667
- Sareena, C., Ramesan, M. T., & Purushothaman, E. (2012). Utilization of coconut shell powder as a novel filler in natural rubber. *Journal of Reinforced Plastics and Composites*, 31, 533-547. doi:10.1177/0731684412439116

- Sarki, J., Hassan, S. B., Aigbodion, V. S., & Ogheneweta, J. E. (2011). Potential of using coconut shell particle fillers in eco-composite materials. *Journal of Alloys and Compounds*, 509(5), 2381-2385. doi:<https://doi.org/10.1016/j.jallcom.2010.11.025>
- Swapna, V. P., Stephen, R., Greeshma, T., Sharan Dev, C., & Sreekala, M. S. (2016). Mechanical and swelling behavior of green nanocomposites of natural rubber latex and tubular shaped halloysite nano clay. *Polymer Composites*, 37(2), 602-611. doi:10.1002/pc.23217
- Wongsorat, W., Suppakarn, N., & Jarukumjorn, K. (2014). Effects of compatibilizer type and fiber loading on mechanical properties and cure characteristics of sisal fiber/natural rubber composites. *Journal of Composite Materials*, 48(19), 2401-2411. doi:10.1177/0021998313498790