Improvement of Hardness Properties of Atural Microfibers: Cotton, Kapok, and Banana as Filler-reinforced Natural Rubber Composites

Pattharaporn Joongpun^{1,*}, Kriangsak Khiaomang¹ and Miyoung Seo¹

¹ Faculty of Fine and Applied Art, Burapha University, Chonburi, 20131, Thailand

Received: 3 December 2022, Revised: 28 December 2022, Accepted: 29 December 2022

Abstract

This work studied the development of mechanical strength and hardness properties of composite natural rubber with natural microfibers of cotton, kapok, and banana at various compositions. The effect of banana fiber length has been studied. The mixed natural microfibers in the natural rubber have been formulated in the proportion of 1, 5, 10, and 15 phr, respectively. The hardness tends to increase in proportion to the natural fiber composite. The hardness measurement results found that the formula MCKB5 was obtained by mixing the fibers. Cotton: Kapok: medium-sized bananas in the proportion of 15 phr in latex had the highest hardness of 76.10 ± 3.19 . The hardness distribution of Shore A shows the uniformity of cotton and kapok fibers. In addition, cotton and kapok fibers promoted the dispersion of banana fibers differently in the case of tri-fiber blends. Rubber composites with natural microfibers under such ideal conditions can improve strength and mechanical properties for reinforced material applications.

Keywords: Natural rubber; Cotton fibers; Kapok fibers; Banana fibers; Shore A hardness

1. Introduction

Natural rubber or para rubber is an important industrial crop in Southeast Asian areas [1]. Typically, previous works in natural rubber fields had focused on developing rubber utilization and processing [2]; each process requires large-scale industrial production tools. It was found that high production was expensive that required intense production skills and technology. Therefore, rubber farmers in the country make it difficult to use technology to improve the value of their rubber products [3]. Moreover, composited rubber material has been received as attractive due to its many advantages such as mechanical properties, low-cost, simple process, etc. [4]-[6]. Usually, composite materials consist of two main components: host matrix and filler materials. The properties of composite material depended on the physical factors of the host matrix and the filler mixer, such as the mixing ratio and the properties filler mixer [6], [7].

Therefore, the other works of natural fibers, local raw materials, were studied to be used as reinforcing materials mixed with natural rubber. It increases the strength properties of natural rubber [8], [9]. Reinforced material from natural fibers is commonly found in many natural sources [10]. Different properties in each fiber type can modify the prepared material's strength. Natural fibers, which have reinforcing properties, can be compared with other fibers [11], [12]. It is another alternative for developing rubber for processing into products that can add value and increase the potential of raw materials in terms of being environmentally friendly [10], [13].

The natural fibers used as reinforcing materials in rubber found that natural fibers can be categorized into four types: stiff fibers, seed fibers (surface fibers), bark fibers (soft fibers),

and animal fibers, which are protein types [14]-[17]. These raw materials are easily found locally throughout the country. Therefore, the authors have selected the fibers specified in this work scope by using cotton fibers with long and flexible fibers, kapok fibers with good absorbency properties, and fibers from bananas with high yield strength as a rubber compound material time [18]-[22]. Based on each fiber's advantages, it is combined to provide the desired formability, filler dispersion, and mechanical qualities.

In this work, three types of natural microfibers (cotton, kapok, and banana sheath) were used as filler materials to increase the strength properties of composited rubber. The physical and mechanical properties of composited rubber were performed. The analyzed results of composite rubber can be used to apply in the efficient processing of rubber products.

2. Experimental

2.1 Preparation of the rubber composites

The banana fiber was initially chosen for this experiment because it was a small, evenly scattered fiber [23], and the cotton fibers effectively adsorbed the natural rubber. Kapok fibers, which are smaller and shorter than cotton fiber and are used in this case, act to absorb natural rubber; they have distributed very well, and the rubber sheet's a steady shape after drying. The photographs and morphology of natural microfibers as shown in Fig. 1.

The fibers were washed with deionized water and dried. They were then dried at 70°C for 3 h and under an atmosphere for 48 h. By cutting with scissors, banana fibers were classified into short, medium, and long categories, with fiber lengths of 2, 6, and 10 mm, as shown in Fig. 2. To explore the impact of banana fiber length, which banana fibers can prevent the tangling of cotton and kapok in the composite rubber. Cotton, kapok, and banana fibers are combined in a blender to begin mixing the fibers before being added to natural rubber during the in-mold process.

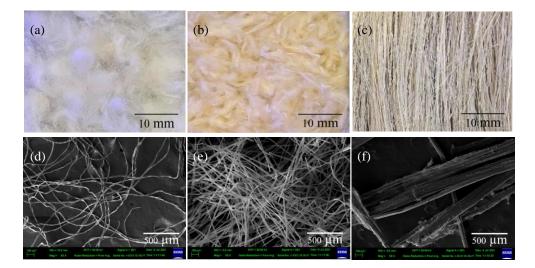


Fig. 1. Photographs and morphologies images of (a)-(d) cotton, (b)-(e) kapok, and (c)-(f) banana, of natural microfibers, respectively



Fig. 2. Photograph images of banana microfiber at various lengths of (a) short (2mm), (b) medium (6 mm), and (c) long (10 mm)

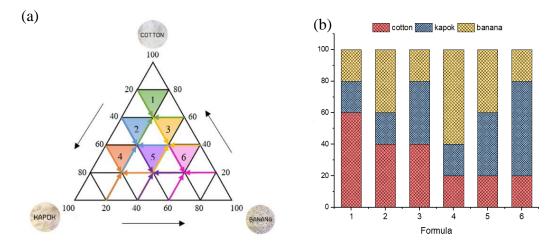


Fig. 3. (a) Triaxial diagram and (b) the proportion of different fiber types in various formulas in this experiment

The different categories of fiber prepared are summarized in Table 1. For the proportion of fibers determined from the triaxial blend diagram of 6 points to find the ratio of 3 types of fibers used for 6 points, the sum of each formula is 100 % by using in Fig. 3. described as follows.

The composition of rubber composite material with a proportion of cotton, kapok, and banana fibers as category names are shown in Table 1. of 1, 5, 10, and 15 (part per hundred of rubber: phr), respectively. For details of raw materials and formulations for preparing rubber composites, the chemicals listed are shown in Table 2. It consists of natural rubber, potassium hydroxide as a gelling agent, zinc dimethyl dithiocarbamate as a catalyst and vulcanizer, butylated reaction product of p-cresol and dicyclopentadiene as an antioxidant, and sulfur as a vulcanizing agent, all of which were purchased at the Rubber Research Institute of Thailand Rubber Authority of Thailand. Zinc oxide nanoparticle (size < 500 nm) is used as a cure activator in the vulcanization process [24], [25].

Type of	Fiber category as fiber proportion (cotton: kapok: banana)						
Banana fiber length	60:20:20	40:20:40	40:40:20	20:20:60	20:40:40	20:60:20	
Short (2 mm) Medium (6 mm) Long (10 mm)	SCKB1 MCKB1 LCKB1	SCKB2 MCKB2 LCKB2	SCKB3 MCKB3 LCKB3	SCKB4 KCKB4 LCKB4	SCKB5 MCKB5 LCKB5	SCKB6 MCKB6 LCKB6	

Table 1. Different natural microfiber composite rubber categories

Table 2. Chemical composition of the natural rubber composite with natural fibers

Ingredient	Content (phr)	
Natural rubber	100	
Potassium hydroxide	0.2	
Zinc dimethyl dithiocarbamate	1.5	
Butylated reaction product of p-cresol and dicyclopentadiene	1	
Zinc oxide nanoparticle	1.5	
Sulfur	2.5	
Natural microfibers (banana: cotton: kapok)	1,5,10,15	

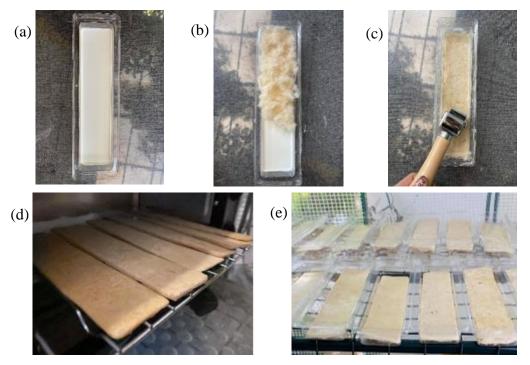


Fig. 4. Preparation of composite rubber: (a) 20% latex is poured, (b) fiber is added to latex, (c) roller is crushed, and the rest of the latex is added; (d) cured at 70 and (e) cool down

For each chemical component that improves mechanical properties, the factor of proportion in rubber that affects mechanical properties was studied. In the case of potassium

hydroxide, the higher the concentration, the higher the strength [26]. For zinc oxide nanoparticles increased, concentration can improve hardness and modulus [24].

The preparation process of composite rubber with natural microfibers at various chemical compositions is shown in Table 2. All fibers in Table 1 were mixed with natural rubber (latex) into a 4.5 x 20 cm mold containing the following chemical composition at the ratio of 1, 5, 10, and 15 phr, respectively. Then, the natural microfibers were added and pressed with a roller, and the remaining latex was added for 1 hour. Next, the composited material was heated to 70°C for 30 minutes in a thermal oven and left to cool down at ambient conditions. Fig. 4 depicts the preparation process of composite rubbers.

2.2 Characterization testing

The properties of composite rubber, such as morphologies and mechanical properties, were performed with scanning electron microscopy (SEM, Zeiss, EVO 15), Shore A hardness test with a durometer with a measuring range between 0 to 100 HD. The hardness number was evaluated under a 5-second indentation time. The median value was recorded from all measured values are 20 locations. A universal testing machine (Instron 4464) was used to measure the stress-strain curves following the ASTM D 412 (Die C) standard for determining tensile strength and elongation at break.

3. Results and discussion

The morphological images of natural microfibers were performed with scanning electron microscopy, as shown in Fig. 5. The results from SEM images were analyzed using the ImageJ program by measuring 20 times. The direct estimation of diameters of cotton, kapok, and banana microfibers are 9.87 ± 3.86 , 21.38 ± 6.77 , and $75.73\pm4.85 \mu m$, respectively. It can be observed that the diameter of banana fibers has decreased from the original because when cotton and kapok fibers are spun, a force acts on the fiber's surface, producing a separation of some of the banana fibers from one another. Fig. 5(d) depicts the morphology of the spun-together fiber.

The hardness of composite rubber under all conditions is greater than 50, as shown in Fig. 6, whereas the hardness of non-fiber composites is 27.62 ± 1.20 . The blend of natural microfibers into the rubber can be seen, resulting in an increased hardness of the material. The hardness ranges from 53.55 ± 4.31 to 73.55 ± 4.51 for formulations using short banana fibers, 52.58 ± 4.29 to 76.61 ± 3.19 for medium, and 52.98 ± 7.09 to 70.30 ± 3.27 for long. The hardness tends to increase when there is a higher proportion of fiber in the rubber composition [6], [27]. However, plotting the data distribution to analyze the variance from the standard deviation, as shown in Fig. 7, allows statistical data to select a suitable formulation for improving the hardness properties of rubber composites. It was found that in the formulations of cotton, kapok, and medium-length banana microfiber (MCKB), there was a slight variance compared to other conditions. As a hardness result of the trend to increase the proportion of fiber blends at high (15 phr), a comparative test was performed in the case of a single natural fiber composite.

From the optimum composite conditions, in the case of 6 mm medium-sized banana fibers, which gave the most outstanding hardness at 15 phr fiber blending. The comparison experiment was repeated in the case of single fibers (cotton, kapok, banana), as shown in Fig. 8. It was found that when mixing the primary single fibers, all fibers could increase the hardness of the rubber, with the highest hardness being 65.65 ± 5.33 from kapok fiber. When the

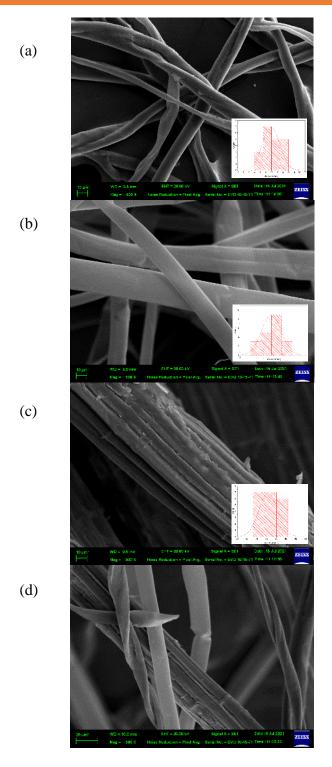


Fig. 5. Morphological images of natural microfibers of (a) cotton, (b) kapok, (c) banana, and (d) mixing of three types of microfibers

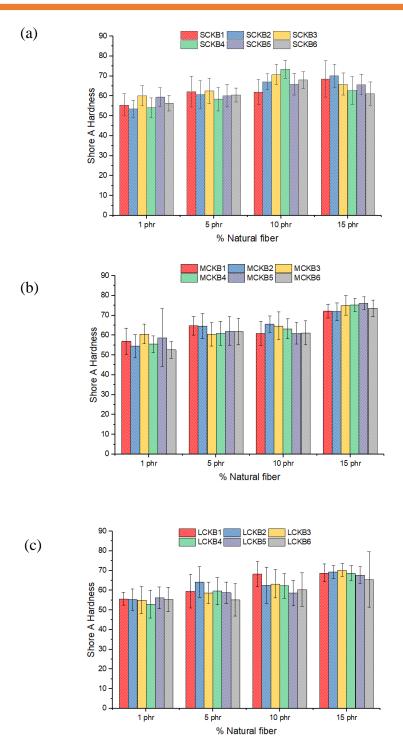


Fig. 6. Hardness values of composite rubber in case of (a) cotton:kapok: short banana fiber (SCKB), (b) cotton: kapok: medium banana fiber (MCKB), and (c) cotton:kapok: long banana fiber (LCKB)

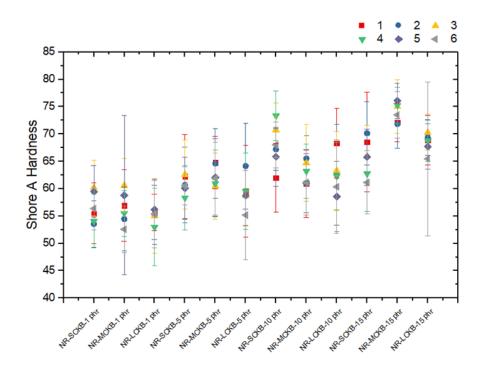
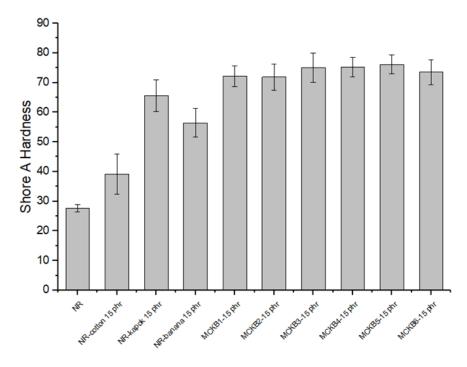


Fig. 7. Shore A hardness of composite rubber



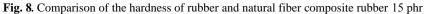
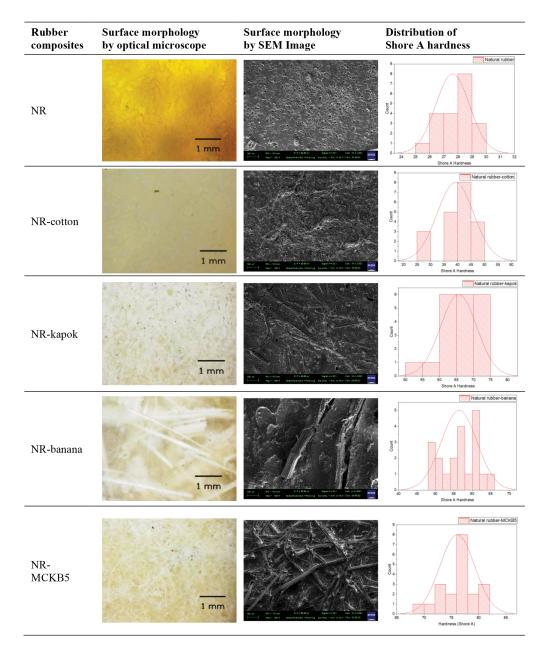


Table 3. Surface dispersion characteristics and Shore A hardness of natural microfibers composite rubber.



three types of fibers were combined, they increased the hardness even more by the examining surface morphology and hardness distribution, as shown in Table 3.

The surface image demonstrates the good compatibility between natural fibers and latex and the homogeneity of cotton, kapok, and blended fibers. The distribution of hardness measured on a rubber sheet, regular natural rubber has a good dispersion since it has a typical rubber texture. When banana fibers were used singly, the hardness measurement may have an unfavorable distribution because the banana fiber cannot be evenly distributed throughout the rubber. However, when three fibers were mixed, the rubber had a consistent distribution of fibers. The banana fibers are dispersed when they coexist because they stick together. It was discovered that the banana fibers' reduced diameter happened during the fiber mixing procedure before adding the natural rubber. Typically, it is well known that composite material's mechanical properties are related to physical properties such as surface roughness or thickness. The decreasing modulus was found when composite film surface roughness increased [28], [29].

According to the study tensile curve in Fig. 9, the pristine rubber composite sheet elongated was 723%. In contrast, composite rubber for NR-Cotton, NR-Kapok, NR-banana, and MCKB5 demonstrated elongation values of 65.2 %, 4.8 %, 524.1 %, and 16.2 %, respectively. In banana fibers, rubber composites have high toughness and high strain, which means that certain rubber parts will continue and retain the characteristics of natural rubber. When it comes to hardness, although kapok is more challenging than other fibers, it was found to have the lowest toughness in all cases, and the case of cotton showed good yield strength compared to kapok and banana and better tensile strain than the case of cotton fibers. However, when using the combination of these three fibers to determine the material's hardness characteristics (MCKB5 formula) can improve the hardness and yield strength, it still found that the toughness was inferior to cotton and banana. It was considering safety applications in the case of rubber composites being used to coat or cover materials to absorb and dissipate energy for impact protection. The main factor is hardness and strength. It can be seen that the rubber composite material developed from the MCKB5 formula is more suitable for use than other fibers. It should be used that focuses on the use that is not very flexible. It is clear that adding natural fibers can increase the hardness of the rubber; however, on the other hand, the elongation is reduced [7], [9].

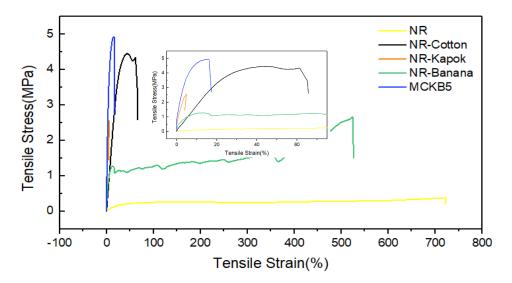


Fig. 9. Tensile curve of natural microfibers composite rubber, inset: the zoom-in of the curve for MCKB5, NR-Kapok, and NR-Cotton

4. Conclusion

In summary, the cotton kapok and banana microfibers were used as reinforcing material for composite rubber, raw materials abundant in the country, to benefit by introducing the microfibers in each composite material. From the results of the natural microfiber blending, it was found that banana fibers were added to cotton and kapok fibers, in addition to helping to break down all fibers well and not clump. From the size of the banana fiber, which has a diameter of 75.73 ± 4.85 micrometers, which is bigger than the two other types, when composited, it caused the insertion between cotton fibers with fiber size 9.87±3.86 and kapok fiber with fiber size 21.38 ± 6.77 µm. Using fiber as a reinforcing material in rubber can improve the hardness properties and yield strength in all fiber cases. In addition, it blends fibers in many conditions and improves them in the optimum conditions. The MCKB5 formula, which combines 6 mm length of banana fibers with the cotton: kapok: banana 20: 40: 40 ratios at the proportion of 15 phr in natural rubber, which has a hardness of 76.1±3.19, was the most suitable considering all the variances of the formula, which is higher than non-fiber composite rubber with a hardness of 27.62±1.20. It was found that the three types of natural fiber composite rubber had a good dispersion effect on the rubber texture and could increase the hardness of composite rubber, and when measured for tensile test, found the highest yield strength. Moreover, it can be processed into various types of products as appropriate, consistent with the properties of the composite rubber of each formula.

Acknowledgment

The authors acknowledge the facilities support from the Faculty of Fine and Applied Art, Burapha University.

References

- Chiarelli DD, Passera C, Rulli MC, Rosa L, Ciraolo G, D'Odorico P. Hydrological consequences of natural rubber plantations in Southeast Asia. Land Degrad. Dev. 2020;31(15):2060-73.
- [2] Bhattacharjee A, Bhowmik M, Paul C, Chowdhury BD, Debnath B. Rubber tree seed utilization for green energy, revenue generation and sustainable development-A comprehensive review. Ind. Crops Prod. 2021;174:114186.
- [3] Simon JN, Nuthammachot N, Techato K, Okpara KE, Channumsin S, Kaewthongrach R, *et al.* Para Rubber (Hevea brasiliensis) Feedstock for livelihoods opportunities in Southern Thailand: Analysis of socioeconomic productivity potentials and security. Sustainability. 2022;14(16):10142.
- [4] Jaafar CA, Zainol I, Ishak N, Ilyas R, Sapuan S. Effects of the liquid natural rubber (LNR) on mechanical properties and microstructure of epoxy/silica/kenaf hybrid composite for potential automotive applications. J. Mater. Res. Technol. 2021;12:1026-38.
- [5] Goyat V, Ghangas G, Sirohi S, Kumar A, Nain J. A review on mechanical properties of coir based composites. Mater. Today Proc. 2022;62:1738-45.
- [6] Roy K, Debnath SC, Pongwisuthiruchte A, Potiyaraj P. Recent advances of natural fibers based green rubber composites: Properties, current status, and future perspectives. J. Appl. Polym. Sci. 2021;138(35):50866.

- [7] Santhosh N, Selvam S, Reghu R, Sundaran J, Mathew BC, Palanisamy S. Mechanical properties studies on rubber composites reinforced with Acacia Caesia fibre. Mater. Today Proc. 2022.
- [8] Jose S, Thomas S, Jibin K, Sisanth K, Kadam V, Shakyawar D. Surface modification of wool fabric using sodium lignosulfonate and subsequent improvement in the interfacial adhesion of natural rubber latex in the wool/rubber composites. Ind. Crops Prod. 2022;177:114489.
- [9] Sivasubramanian P, Mayandi K, Santulli C, Alavudeen A, Rajini N. Effect of fiber length on curing and mechanical behavior of pineapple leaf fiber (PALF) reinforced natural rubber composites. J. Nat. Fibers 2022;19(11):4326-37.
- [10] Karimah A, Ridho MR, Munawar SS, Adi DS, Damayanti R, Subiyanto B, *et al.* A review on natural fibers for development of eco-friendly bio-composite: Characteristics, and utilizations. J. Mater. Res. Technol. 2021;13:2442-58.
- [11] de Azevedo AR, Cruz AS, Marvila MT, de Oliveira LB, Monteiro SN, Vieira CMF, *et al.* Natural fibers as an alternative to synthetic fibers in reinforcement of geopolymer matrices: a comparative review. Polymers. 2021;13(15):2493.
- [12] Gupta M, Ramesh M, Thomas S. Effect of hybridization on properties of natural and synthetic fiber-reinforced polymer composites (2001–2020): A review. Polym. Compos. 2021;42(10):4981-5010.
- [13] Rajeshkumar G, Seshadri SA, Devnani G, Sanjay M, Siengchin S, Maran JP, et al. Environment friendly, renewable and sustainable poly lactic acid (PLA) based natural fiber reinforced composites-A comprehensive review. J. Cleaner Prod. 2021;310:127483.
- [14] Ganguly A, Shankar S, Das A, Shukla M, Swaroop C, Bhardwaj T. Natural fibre reinforced composites: A review based on additive manufacturing routes and biodegradability perspective. Mater. Today Proc. 2022.
- [15] Guru R, Kumar A, Kumar R. Basic Functional application for natural fibers and types. In: Han-Yong Jeon. Natural fibers. 1st ed. Intech Open; 2022. p.1-14.
- [16] Kumar GCM, Mallik S. Natural fibre-reinforced polymer composites. In: Sultan MTH, Rajesh M, Jayakrishna K. Failure of fibre-reinforced polymer composites. 1st ed. Boca Raton: CRC Press; 2021. p. 1-11.
- [17] Karimah A, Ridho MR, Munawar SS, Amin Y, Damayanti R, Lubis MAR, et al. A comprehensive review on natural fibers: Technological and socio-economical aspects. Polymers. 2021;13(24):4280.
- [18] Boppana SB, Palani Kumar K, Ponshanmugakumar A, Dayanand S. Different natural fiber reinforced composites and its potential industrial and domestic applications: A review. In: Palanikumar K, Thiagarajan R, Latha B. Bio-fiber reinforced composite materials. 1st ed. Springer link; 2022. p. 51-73.
- [19] Rangappa SM, Siengchin S, Parameswaranpillai J, Jawaid M, Ozbakkaloglu T. Lignocellulosic fiber reinforced composites: Progress, performance, properties, applications, and future perspectives. Polym. Compos. 2022;43(2):645-91.
- [20] Yadav V, Singh S. A comprehensive review of natural fiber composites: Applications, processing techniques and properties. Mater. Today Proc. 2021.
- [21] Prakash SO, Sahu P, Madhan M, Johnson Santhosh A. A review on natural fibrereinforced biopolymer composites: properties and applications. Int. J. Polym. Sci. 2022;7820731.

- [22] Kumar RG, Rajesh DR. A study on the abrasion resistance, compressive strength and hardness of banana–fibre reinforced natural rubber composites. IJARET 2016;7(3):42-55.
- [23] Deepa C, Ramesh M. Banana fibers, their composites and applications. Plant Fibers, their composites, and applications: Elsevier; 2022. p. 161-80.
- [24] Hadi F, Kadhim R, editors. A study of the effect of nano zinc oxide on cure characteristics and mechanical properties of rubber composites. J. Phys. Conf. Ser. 2019;1234:012043.
- [25] Sreethu TK, Naskar K. Zinc oxide with various surface characteristics and its role on mechanical properties, cure-characteristics, and morphological analysis of natural rubber/carbon black composites. J. Polym. Res. 2021;28(5):1-14.
- [26] Sutanto TD, Setiaji B, Wijaya K, Suharto TE. Effect of KOH as stabilizer on mechanical and chemical properties of liquid rubber compound. Asian J. Chem. 2014;26(24):8371.
- [27] Gairola S, Tyagi Y, Gangil B, Sharma A. Fabrication and mechanical property evaluation of non-woven banana fibre epoxy-based polymer composite. Mater. Today Proc. 2021;44:3990-6.
- [28] Kwaśniewska A, Świetlicki M, Prószyński A, Gładyszewski G. Physical properties of starch/powdered activated carbon composite films. Polymers. 2021;13(24):4406.
- [29] Vilela MS, Bernal VL, Chagas LLC, Vichi FM, Aranha ACC, Arana-Chavez VE, et al. Mechanical properties and surface roughness of polymer-based materials containing DCPD particles. Brazilian Oral Research. 2020;34.