



Enhancing natural rubber properties: a comprehensive study on the synergistic effects of wood sawdust and carbon black as fillers in rubber composites

İlyas Kartal¹ · İdris Karagöz²

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Abstract

Rubber products, especially in the automotive industry, play a vital role in manufacturing. However, natural rubber initially has relatively low mechanical properties. To enhance these properties, various additives and fillers are incorporated, including inorganic and organic options. This study focused on investigating the mechanical and thermomechanical properties of natural rubber composites filled with wood sawdust and carbon black. Tests such as hardness, density, tensile strength, viscosity, tear resistance, rebound resilience, accelerated aging, rubber process analysis, and dispersion were conducted to assess the viability of using wood sawdust as a filler material. The results indicate that wood sawdust, in combination with carbon black, can effectively serve as a filler in natural rubber when the appropriate filling ratio and size are carefully selected to achieve the desired properties. These findings underscore the potential of utilizing wood sawdust as a sustainable and cost-effective filler option in the production of rubber materials, especially in the automotive industry. Further research and optimization efforts have the potential to contribute significantly to the development of high-performance rubber composites with enhanced mechanical characteristics.

Keywords Natural rubber composites · Wood sawdust · Natural filler · Mechanical properties · Tensile strength

✉ İlyas Kartal
ilyaskartal@marmara.edu.tr

✉ İdris Karagöz
idris.karagoz@yalova.edu.tr

¹ Metallurgy and Material Engineering, Marmara University, 34854 Istanbul, Turkey

² Polymer Materials Engineering, Yalova University, 77200 Yalova, Turkey

Introduction

Natural rubber (NR) composites, known for their exceptional mechanical properties and biodegradability, have emerged as eco-friendly alternatives to traditional materials [1]. To enhance their performance, researchers have explored reinforcing agents such as wood sawdust, a cost-effective byproduct of wood processing. Incorporating sawdust into NR composites not only addresses waste concerns in the wood industry but also leads to improved composite properties, offering a sustainable solution [2].

Recent research efforts have concentrated on enhancing the mechanical properties of polymer composites. Specifically, there has been a focus on cellulose-based fibers derived from both wood and non-wood sources [2, 3]. These fibers offer environmental benefits, cost-effectiveness, and favorable performance traits. With attributes like lower density, high specific strength, and stiffness, they enable the production of low-density composites with higher filler concentrations. Their flexibility reduces the risk of fracture during processing, making them highly desirable for composite applications. Natural fiber-reinforced polymer composites also offer advantages such as low energy costs, positive environmental impact, biodegradability, recyclability, and excellent thermal properties, making them attractive for various applications [3].

Researchers are actively developing biodegradable "green" composites by blending natural fibers with rubber, aiming for environmentally friendly materials that can be easily disposed of without harming the environment. These fiber-reinforced rubber composites play a crucial role in various applications, combining a soft, elastic rubber matrix with a strong, stiff fibrous reinforcement [4, 5]. They outperform aluminum in hardness and steel in stiffness, making them highly valuable in industrial fields. Short fiber-reinforced rubber composites, in particular, have gained popularity due to their processing advantages and enhancements in strength, stiffness, modulus, and damping [4–9]. Designing these composites requires careful consideration of factors such as fiber aspect ratio, control of fiber orientation and dispersion, and the establishment of a strong fiber-rubber interface [3]. Ongoing research in this area is vital for creating innovative materials that meet both industry demands and environmental sustainability requirements.

In rubber materials, a complex relationship arises between the size of the organic-based filler and the properties it imparts to the structure. This relationship can vary depending on the type of filler material, its distribution and other factors. Generally, the size of the filler material affects the interfacial area between the matrix and the filler. Small-sized fillers can provide a stronger bond with the matrix due to their larger interfacial area, which can increase the mechanical strength of the material. However, excessively small fillers can be difficult to distribute homogeneously, potentially negatively affecting the overall performance of the material. On the other hand, larger-sized fillers have a smaller interfacial area and may provide lower mechanical strength, but they can be advantageous in terms of processability and production costs. A review of the

literature reveals that different filler sizes are preferred for various applications, such as enhancing the mechanical properties in tire manufacturing [10], improving thermal stability in seals and gaskets [11], and reducing production costs in bulk rubber goods [12].

Numerous studies have been conducted on natural fibers and/or fillers. The literature includes many investigations, such as the effect of sawdust in thermoplastics [13] and the effect of sawdust size in thermosetting materials [14]. Similar studies have been conducted on rubbers. Kaewpruk et al. [15] investigated the use of 45 μm sawdust and wood ash as fillers in natural rubber composites. Ahmad et al. used sawdust with a size of 100–200 μm in their mechanical and morphological studies of recycled PET-based rubber wood sawdust filled UPR composites [16]. Shakir et al. used sawdust particles with an average size of 686 μm in their study on the conversion of rubber wood sawdust to green mycelium-based composite [17]. Manaila et al. produced wood sawdust/natural rubber eco-composites using sawdust with a size of 50 μm [18]. Sombatsompop et al. [19] used sawdust sizes of 100–300 μm in their study on wood sawdust fibers as secondary fillers in carbon black-filled NR vulcanizates. As can be seen, the sawdust size used in the studies varies. In this study, waste wood sawdust in sizes of 0–200 μm , 200–400 μm , and 400–600 μm was preferred.

Various natural fibers, including sisal, coir, waste silk, jute, and bamboo, have been explored to reinforce natural rubber composites [20–25]. Despite the abundance of waste wood sawdust in the wood processing industry, research on natural rubber and waste wood sawdust remains limited [26, 27]. Delving deeper into this field, conducting further research, and intensifying efforts are crucial. Combining natural rubber with waste wood sawdust has the potential to offer both environmental and economic advantages. Utilizing waste wood sawdust as a filler material in green composites, along with natural rubber's excellent properties, provides a dual benefit. This combination enhances natural rubber properties and significantly contributes to waste management efforts. Studying natural rubber and waste wood sawdust facilitates the development of eco-friendly and sustainable materials, promoting sustainability in industrial processes. Advocating for and supporting research in this promising area is imperative to advance eco-friendly materials and practices.

The study aimed to assess natural rubber composites reinforced with waste wood sawdust fillers, incorporating various additives to enhance rubber properties. The mixtures underwent multiple tests, including hardness, density, tensile strength, Mooney viscosity, tear resistance, rebound resilience, and permanent deformation, with additional aging tests conducted for material stability. Dynamic properties were measured using a rubber process analyzer, and filler dispersion was analyzed. While the study focused on evaluating waste wood sawdust's suitability as a reinforcing filler for natural rubber composites, emphasizing mechanical properties over morphological investigations, future research plans to explore these features as a separate focus, recognizing their potential for pioneering advancements in the field.

Materials and methods

Materials

The rubber samples were prepared using natural rubber (SVR 3L, Vietnam) as the matrix material. Various additives were incorporated to enhance the rubber's properties and achieve the desired performance. The fillers included waste wood sawdust in three different sizes (0–200 μm , 200–400 μm , and 400–600 μm) at 40 parts per hundred rubber (PHR) and carbon black (N550, pH 6–8, ash content 0.75%) at 40 PHR. Sulfur (S 80) at 2.5 PHR served as the curing agent, while vulcanization acceleration was achieved with 0.6 PHR CBS 80 (density 1.05 g/cm^3 , Cyclohexyl-2-benzothiazolesulfenamide). Additionally, 0.1 PHR CTP 80 (density 1.23 g/cm^3 , 80% N-(cyclohexylthio)-phthalimide, 20% elastomer binder, and dispersing agents) was used as the retarder. To facilitate sulfur activation, 5 PHR zinc oxide was added. Softening was accomplished with 1 PHR stearic acid. Antioxidants, namely 4010 (IPPD) and TMQ (polymerized 2,2,4-trimethyl-1,2-dihydroquinoline), along with microcrystalline wax, were included. Processing additives, Kumaron C100 and Struktol A60, were used at a total of 13 PHR for other chemicals. Additionally, 20 PHR naftenic oil was employed as a lubricant in the process. The mixtures and their corresponding codes are detailed in Table 1. The sample coded as "Natural Rubber (Ref.)" in Table 1 represents natural rubber without any filler. The percentages shown as 0% in the graphs correspond to this filler-free natural rubber.

Preparation of mixtures

To ensure a uniform mixture, the materials underwent a pre-mixing process in a Banbury mixer (Werner & Pfeiderer GK1.5N) in accordance with the ISO 2393 standard. Initially, rubber was introduced into the mixer and mixed for two minutes. Following this initial mixing, protective agents and activators were added to the mixture, and mixing continued until the temperature dropped. Once

Table 1 Wood sawdust ratio and particle sizes in mixtures

Code*	SVR 3L (PHR)	Wood sawdust (PHR)	Wood sawdust particle size (μ)
Natural Rubber (Ref.)	100	–	–
NR + WS10A	100	10	0–200
NR + WS10A	100	20	0–200
NR + WS10A	100	30	0–200
NR + WS10B	100	10	200–400
NR + WS20B	100	20	200–400
NR + WS30B	100	30	200–400
NR + WS10C	100	10	400–600
NR + WS20C	100	20	400–600
NR + WS30C	100	30	400–600

the temperature reached 120 °C, the rubber compound was transferred to an open mill. In the open mill, a cooking chemical was added for one minute with a 2-mm mill gap. Subsequently, the rubber compound underwent six rounds of mixing with a 2-mm mill gap, four rounds with a 3-mm mill gap, two rounds with a 4.5-mm mill gap, and two rounds with a 5-mm mill gap to complete the mixing process. Waste wood sawdust in sizes of 0–200 μm, 200–400 μm, and 400–600 μm was added to the other prepared mixtures during the pre-mixing stage according to the PHR (Parts per Hundred Rubber) ratios specified in Table 1, in addition to the reference sample. The stages from pre-mixing to characterization of the samples in the Banbury mixer are schematically illustrated in Fig. 1. The first section of the figure represents the materials used, the second section shows the mixing stage, and the final section depicts the characterization process.

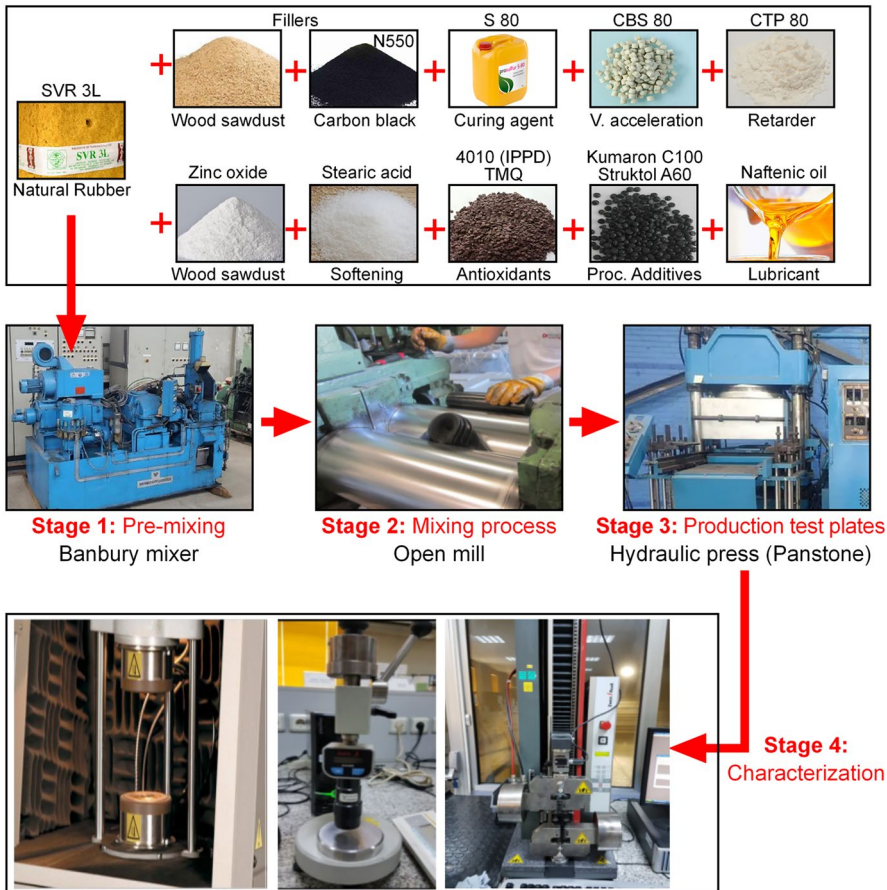


Fig. 1 Schematic representation of the processes from sample preparation to characterization

Production of test plates

Test specimens were obtained from the prepared rubber compounds through a molding process using a hydraulic press (Panstone) in accordance with ISO 2393. The rubber mixtures were placed into molds of standard dimensions and subjected to the vulcanization process by being held in the mold at a temperature of 150 °C and under pressure for the optimal curing time determined by rheometer curves. Following vulcanization, test specimens suitable for the intended tests were cut from the vulcanized plates using standard cutting blades.

Characterization

To assess the properties of the prepared mixtures, various tests were conducted on the test specimens, including hardness, density, tensile strength, Mooney viscosity, tear resistance, resilience, and permanent deformation tests. Additionally, an aging process was performed by exposing the specimens to air at 100 °C for 72 h.

Hardness measurements were performed in Shore A units at room temperature using a Zwick durometer, following the ISO 7619-1 standard. The densities of the specimens were determined with a Sartorius density kit according to the ISO 1183-1 standard and guidelines of ISO 2781. Tensile tests were carried out on S1 type specimens using a Zwick testing machine, adhering to the ISO 37 standard, at room temperature and a crosshead speed of 500 mm/minute.

Mooney viscosity testing was conducted using an Alpha MV2000 device following the ISO 289-1 standard. Tear tests were performed on Method A (trouser) specimens using a Zwick tensile testing machine at room temperature and a crosshead speed of 100 mm/minute, as per the ISO 34–1 standard. Resilience tests were conducted using a Karl Frank GMBH resilience tester following the DIN 5312 standard, with specimens conditioned for 3 h at 23 ± 2 °C. The permanent deformation test was performed according to the ISO 815–1 standard, compressing cylindrical specimens by 25% and aging them in an oven at 100 °C for 72 h.

Accelerated aging tests involved keeping the specimens in a Nuve oven at 100 °C for 72 h, following the DIN 53508 standard. Dynamic properties were measured using an Alpha RPA200 rubber process analyzer (RPA) in accordance with the ASTM D6420-4 standard. Elastomer frequency tests measured complex modulus and $\tan \delta$ values at 40 °C and 7% strain, at frequencies of 1.5, 10, and 20 Hz. Elastomer strain sweep tests measured complex modulus and $\tan \delta$ values at 40 °C and a frequency of 1.7 Hz, at strains of 1%, 10%, 20%, and 40%. Dispersion tests were conducted with a MonTech disperser device according to the ISO 11345 standard, analyzing x, y, and z values, percent dispersion, and white area.

Results and discussion

Hardness and density measurement results

In composite materials, a complex relationship arises between the size of the organic filler and the hardness properties it imparts to the structure. Generally, larger and denser fillers can contribute to a higher hardness level, but this can vary depending on the type of material, its distribution and other factors. The process of combining organic fillers with the composite material can also affect its hardness properties. For instance, the pressing processes used during composite sample preparation can impact the homogeneity of the material and thus its hardness. Additionally, when open casting is performed, the fillers may settle to the bottom due to the density differences. Furthermore, organic fillers can affect the properties of the composite and its hardness due to humidity, temperature, or chemical interactions [28].

The hardness measurement results, presented in Fig. 2a, and the density measurement results, illustrated in Fig. 2b, provide valuable insights into the wood sawdust-filled rubber composites. The unfilled natural rubber specimen exhibited a hardness of 36 Shore A, serving as a baseline reference. The hardness values of the rubber composites varied based on the filler content and particle size. Notably, increasing filler content and decreasing particle size led to higher hardness values. Additionally, an increase in particle size, while maintaining constant filler content, also resulted in elevated hardness values, highlighting the impact of wood sawdust filler on the composite's hardness.

Density measurements revealed similar trends. The density of the unfilled natural rubber stood at 1.073 g/cm^3 , providing a benchmark for comparison. With the incorporation of wood sawdust filler, the density of rubber composites increased proportionally with filler content and particle size. Higher filler content corresponded to increased density, emphasizing the direct relationship between the two. Similarly, particle size variations influenced density, indicating the flexibility to optimize rubber composite density by adjusting filler content and particle size based on specific application requirements.

Notably, the observed increase in density coincided with the rise in hardness, suggesting a positive correlation between density and hardness in these composites. However, it is essential to acknowledge that other factors could influence this relationship. Further in-depth analysis and experimentation are warranted to refine our understanding of the intricate interplay between density and hardness in wood sawdust-filled rubber composites.

Mooney (viscosity) testing results

Mooney viscosity emerges as a pivotal parameter in assessing the processability and vulcanization characteristics of natural rubber (NR). A higher Mooney viscosity value indicates heightened resistance to flow, reflecting an increased

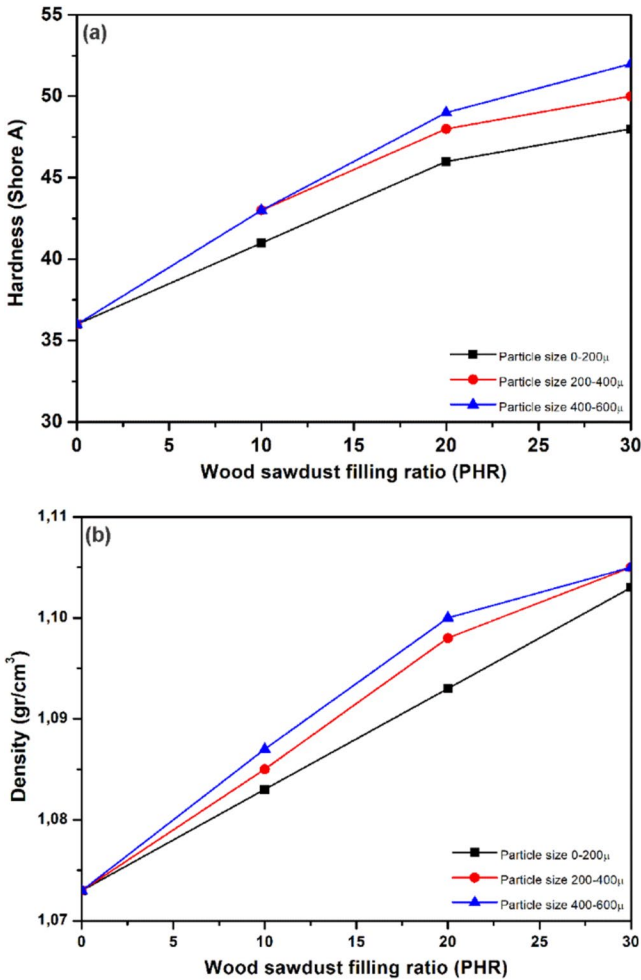


Fig. 2 Hardness and density measurement results **a** hardness **b** density

molecular weight and a more extensive degree of polymerization within the rubber sample [29, 30]. Figure 3a provides a detailed comparison of Mooney viscosity values for different particle sizes, while Fig. 3b focuses on samples within the 0–200 μm range. Figure 3c presents Mooney values for particle sizes ranging from 200 to 400 μm , and Fig. 3d showcases values for particle sizes from 400–600 μm .

Generally, the introduction of wood sawdust filler particles results in an elevation of Mooney viscosity in natural rubber composites. The results distinctly highlight viscosity variations based on filler loading and particle size range. As filler loading or particle size increases, a consistent upward trend in viscosity becomes evident. It is apparent that the inclusion of wood sawdust particles significantly influences the viscosity of the rubber composite. Furthermore, the size and geometry of the wood

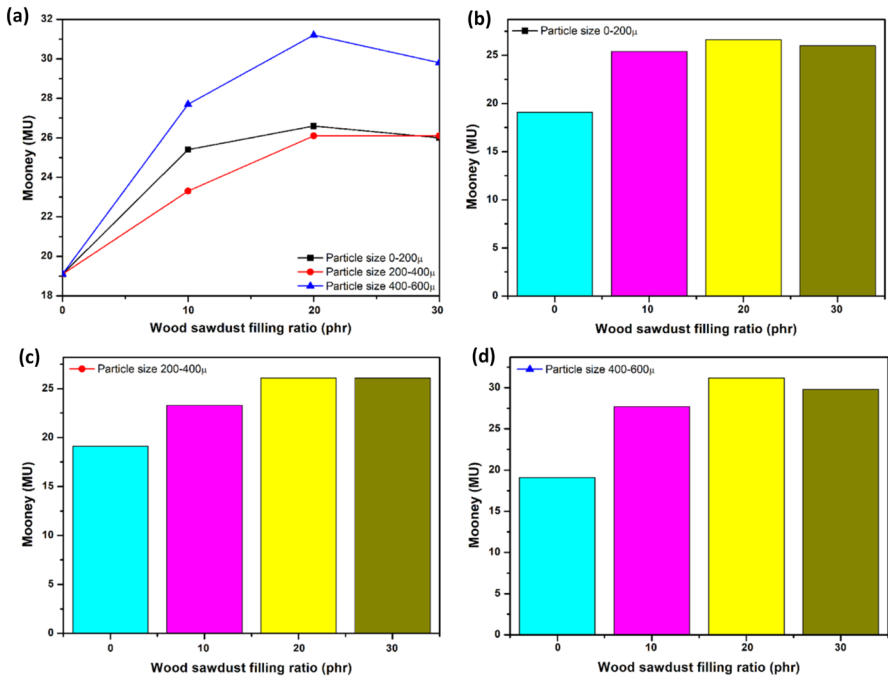


Fig. 3 Comparison of Mooney test results **a** Mooney viscosity for different phr, **b** Mooney viscosity for 0–200 μ particle sizes **c** Mooney viscosity for 200–400 μ particle sizes and **d** Mooney viscosity for 400–600 μ particle sizes

sawdust particles play a crucial role in determining viscosity. Mechanical processing and the duration applied to the sawdust further contribute to these viscosity changes [29]. The strong correlation observed in the Mooney viscosity values among the natural rubber composite samples underscores the significant impact of wood sawdust filler on the rubber’s processability and vulcanization characteristics.

Tensile test results

The comparison of tensile strength values for different particle sizes is visually depicted in Fig. 4a, while the corresponding % elongation values are presented in Fig. 4c. Figure 4b delves into the impact of wood sawdust with varying phr values within their respective particle size ranges, as well as with other particle sizes, on tensile strength in rubber. Figure 4d provides a similar comparison for % elongation value.

Tensile strength, a measure of a material’s maximum stress tolerance before breaking or deforming, was evaluated for the natural rubber composites. The reference natural rubber exhibited a tensile strength of 89 kgf/cm². Introducing 10 phr of 0–200 μ m particle size wood sawdust filler enhanced the tensile strength to 113 kgf/cm², indicating the reinforcing effect of the filler. However, at 20 phr and

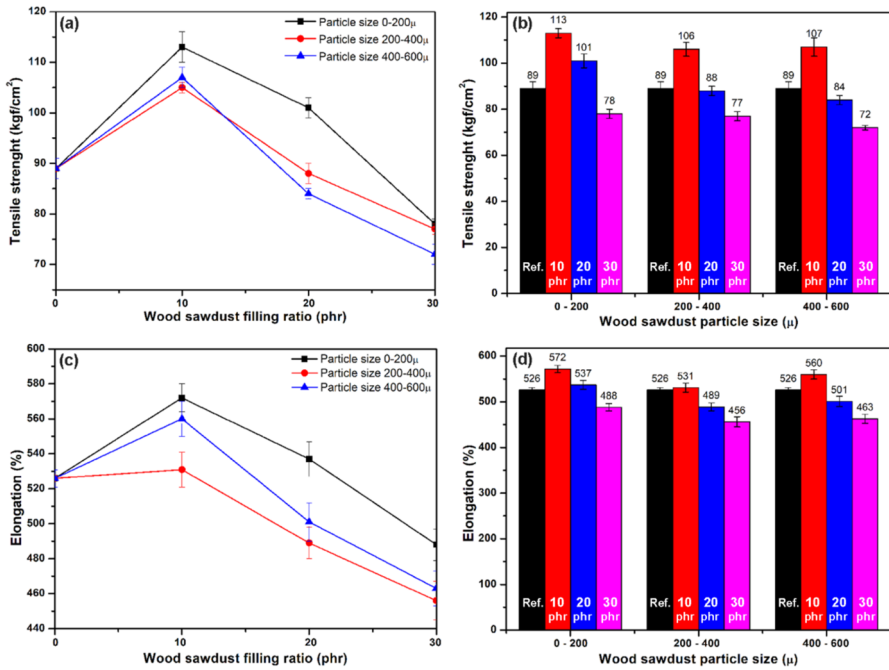


Fig. 4 Tensile test results **a** Comparison of tensile strength for different phr, **b** Comparison of tensile strength for different particle sizes **c** comparison of elongation for different phr and **d** comparison of elongation for different particle sizes

30 phr filler content, the tensile strength decreased to 101 kgf/cm² and 78 kgf/cm², respectively. This suggests the existence of an optimal filler content, beyond which tensile strength decreases. Similar trends were observed for composites filled with 200–400 μ m and 400–600 μ m particle size wood sawdust, where increasing filler content reduced tensile strength. These findings underscore the significant influence of filler type and content on the tensile properties of natural rubber composites [31, 32]. A careful balance of filler content is crucial to achieving the desired mechanical properties while avoiding excessive loading that could be detrimental to tensile strength.

The elongation at break of the reference natural rubber measured 526%. Incorporating 10 phr of 0–200 μ m particle size wood sawdust filler increased elongation to 572%, indicating improved flexibility. However, for composites with 20 phr and 30 phr filler content, elongation decreased to 537% and 488%, respectively. Higher filler contents in composites with 200–400 μ m and 400–600 μ m particle size wood sawdust showed decreasing elongation trends. At 30 phr filler content, elongation at break values was 456% and 463%, respectively. Low filler content can enhance elongation, but higher contents may reduce it. Desired elongation and filler content must be carefully considered in designing natural rubber composites that meet specific elongation requirements [31, 32].

Tear resistance test results

In rubber applications subjected to high strains, such as in automotive or industrial products, the material may experience cracks or tears. The tearing characteristics of a rubber mixture depend on factors like crosslink density, degree of vulcanization, and the type of filler used. The force required to initiate a rupture differs from the force needed to propagate it. Tear tests, often conducted using the trousers sample, are essential for studying crack propagation in rubber. Natural rubber, owing to its inherent structure, exhibits good tear resistance [33–35].

Figure 5 presents the tear test results. The mixtures showed a rapid decrease in tear resistance up to a 20% filler ratio, after which the rate of decrease slowed down. Comparing tear test results based on sawdust sizes revealed that the PHR20 and PHR30 mixtures yielded similar results. In the PHR10 mixture, the decrease in tear strength slowed down as the sawdust size increased. Generally, the addition of wood sawdust to the mixtures led to a decrease in tear resistance. According to the test results, natural rubber, with its inherent tear resistance, can tolerate up to a 50% decrease in tear strength when sawdust is used as a filler in rubber applications. This suggests that wood sawdust can be a viable filler option for rubber compounds, especially when tear resistance requirements allow for such tolerances.

Rebound resilience test results

Figure 6 illustrates the results of the rebound test, a fundamental method for evaluating the dynamic properties of rubber materials. Despite its simplicity, this test provides valuable insights into the characteristics of rubber products, as it correlates with various dynamic properties. Upon analyzing the rebound resilience test results, a partial increase was observed with higher sawdust filling ratios, followed by stabilization. The influence of the sawdust ratio was not significant. When considering sawdust size, partial changes were noted. Smaller sawdust size had a more positive effect, whereas overall size change did not produce a noticeable

Fig. 5 Tear resistance test results

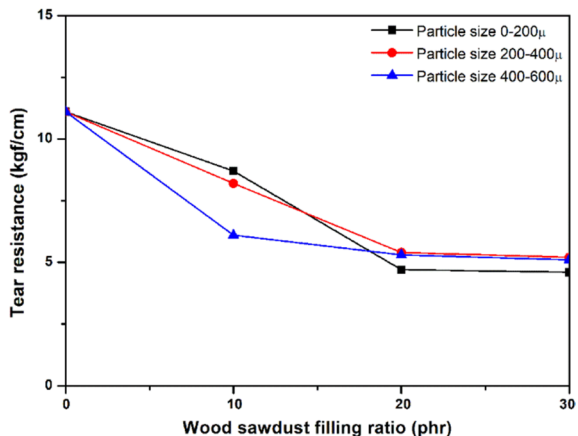
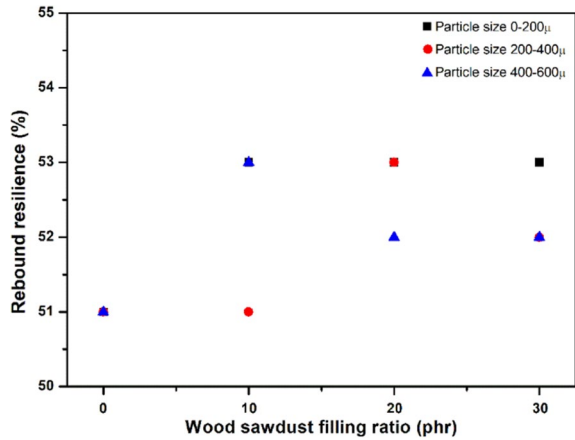
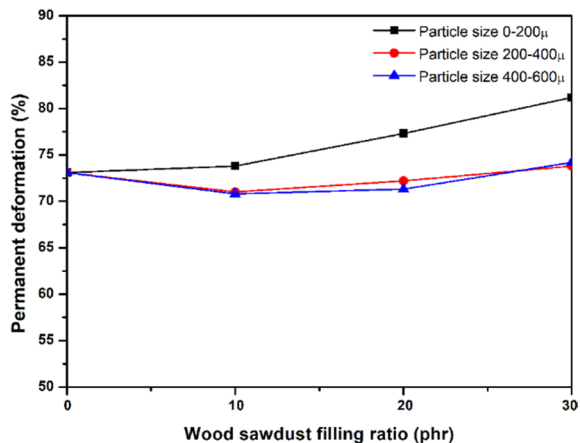


Fig. 6 Rebound resilience rate

impact. These observations highlight the subtle yet impactful role of sawdust filling ratios and particle size in the rebound resilience of rubber composites.

Permanent deformation tests results

The permanent deformation test results depicted in Fig. 7 highlight the variability in permanent deformation based on the sample's shape, preconditioning, and deformation rate [36]. When comparing the permanent deformation test results according to the filler ratio in the mixture, distinct differences emerged in the sample group with 0–200 μ m sawdust size. Permanent deformation increased as the filling rate increased for the 0–200 μ m sawdust size. At 200–400 μ m and 400–600 μ m sawdust sizes, the permanent deformation decreased by up to 10% before showing an increase. Analyzing permanent deformation based on sawdust size, it rose up to 0–200 μ m sawdust size. Then, as the sawdust size increased, deformation

Fig. 7 Permanent deformation rate

decreased, except for the PHR30 mixture. In the PHR30 mixture, a partial increase in deformation values was observed at 400–600 μm . Generally, to minimize deformation in the permanent deformation test, it was concluded that the filling ratio should be kept low, and the sawdust size should be increased. These findings emphasize the importance of careful consideration of filler ratio and particle size to control permanent deformation effectively.

Accelerated aging test results

Natural rubber’s inherent lack of oxidative aging resistance, ozone resistance, UV resistance, and thermal aging performance due to unsaturation on the chain is a well-known challenge [33–35]. To assess the impact of aging, the mixtures were subjected to tensile tests after aging in air at 100 °C for 72 h, and the changes in the data are presented in Fig. 8.

As expected, the tensile strength decreased after aging, a common phenomenon for natural rubber. Interestingly, before aging, all samples at the 10% filling ratio showed an increase in tensile strength. However, after aging, this increase was limited to only the 0–200 μm sawdust size, and the tensile strength after 10% filling was lower than that of the reference sample. Moreover, the tensile strength decreased

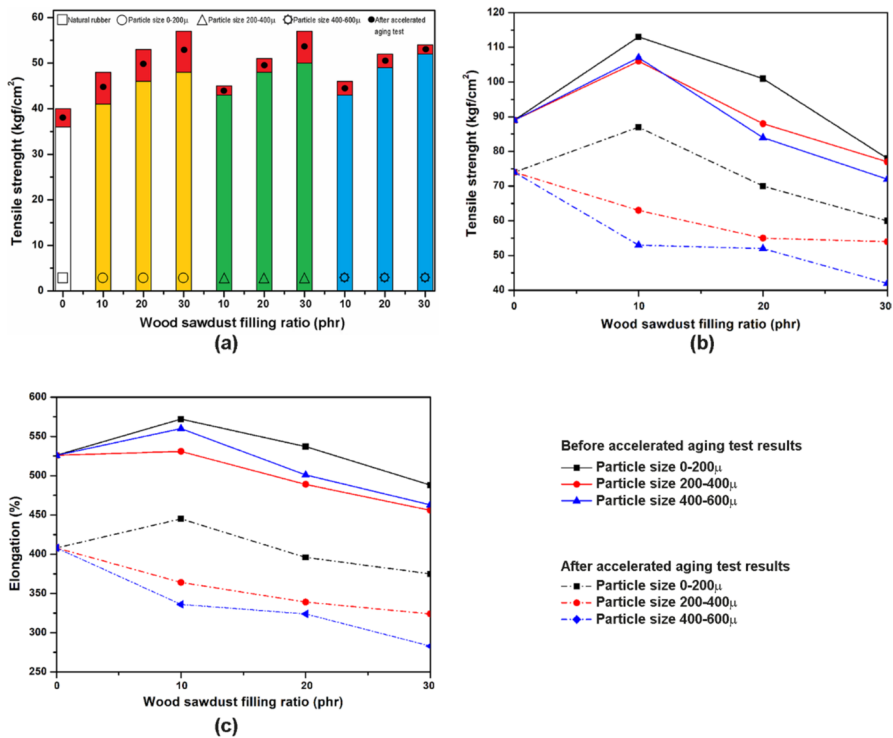


Fig. 8 Accelerated aging test results

with the addition of more filler, and the decrease was more prominent in larger-sized samples, aligning with expectations.

Analyzing the accelerated aging test results, it was observed that to maintain strength after the air aging process, it is advisable to keep the filling ratio low, regardless of the sawdust size. After air aging, the elongation values of the mixtures decreased. The % elongation graph displayed a similar pattern in both pre-aging and post-aging tensile test graphs. A notable increase in % elongation was observed with a 10% filling in all sizes, followed by a subsequent decrease. Interestingly, the tensile strength of the sample without additives at the filler size with 30% addition was found to be below the strength of the additive-free sample. The highest % elongation value was recorded in 10% filled samples. Thin samples consistently exhibited higher elongation across all tested conditions. These results underscore the intricate relationship between filler content, particle size, and aging effects on natural rubber composites, emphasizing the need for careful selection and optimization to achieve desired mechanical properties after aging.

Rubber process analyzer test results

The elastomer frequency scanning results were analyzed across a range of 1–20 Hz, focusing on two key parameters: G^* and $\tan \delta$. The G^* results in elastomer frequency scanning were found to be directly proportional to the hardness values of the mixtures, while $\tan \delta$ values exhibited an inverse relationship with the dynamic values of the mixtures (Fig. 9). Specifically, G^* values at 1 Hz and 20 Hz ranged between 424 and 700 (Fig. 9a), and $\tan \delta$ values ranged from 0.108 to 0.152 (Fig. 9b).

Further examination of elastomer frequency scanning results, specifically G^* and $\tan \delta$ values, was conducted based on the amount of waste sawdust added to the mixture, the filling ratio, and the sawdust size. When comparing results based on filling ratio, an increase in G^* values was observed with higher Hz and filling ratios, with the optimum G^* values occurring at a 30% filling ratio. The sawdust size, especially in the range of 400–600 μm , played a crucial role in achieving the highest G^* values in the mixtures.

In terms of $\tan \delta$ values, a general decrease was noted as the filling ratio increased. However, variations occurred with different sawdust sizes, with a 10% and 20% increase in $\tan \delta$ values for sawdust sizes of 200–400 μm and 400–600 μm , respectively. The trend reversed when reaching a 30% filling ratio, indicating the intricate relationship between $\tan \delta$ values, filling ratio, and sawdust size. Comparisons based on sawdust size revealed that both G^* and $\tan \delta$ values increased with larger sawdust sizes. The increase in G^* values was more pronounced in mixtures with higher filler contents (PHR20 and PHR30) when transitioning from no sawdust to 0–200 μm size, with a subsequent slowing down of the increase. The sawdust size, especially in the 400–600 μm range, was identified as critical for achieving the highest G^* values. In elastomer strain scanning results, G^* values were found to be proportional to the hardness values of the mixtures, while $\tan \delta$ values were inversely proportional to the dynamic values. G^* values ranged between

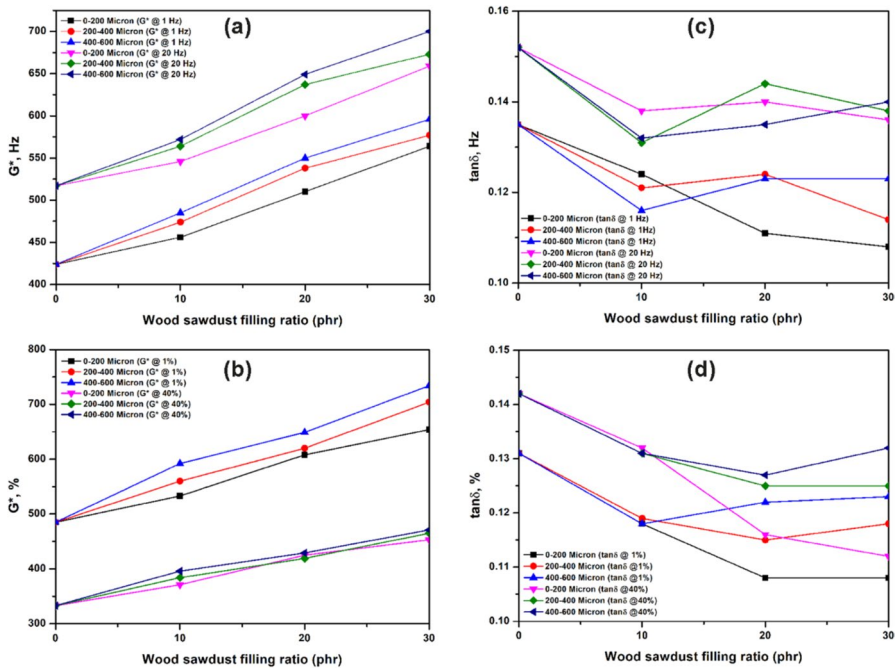


Fig. 9 Rubber process analyzer test results **a** G^* @ 1 Hz and 20 Hz **b** G^* @ 1% and 40% **c** $\tan \delta$ @ 1 Hz and 20 Hz **d** $\tan \delta$ @ 1% and 40%

333 and 734 (Fig. 9c), and $\tan \delta$ values ranged from 0.108 to 0.142 when values were examined at 1% and 40% (Fig. 9d). Analyzing G^* values in elastomer strain scanning revealed an increase with both higher filling ratios and larger sawdust sizes. The highest G^* values were associated with a high filling ratio. When elastomer strain scanning results were analyzed according to sawdust size, an increase in G^* values was observed with larger sawdust sizes, especially when transitioning from 0 to 200 μm sawdust size.

Comparing $\tan \delta$ values with sawdust size indicated a decrease with 0–200 μm sawdust size, followed by an increase with larger sizes, except for the PHR10 mixture. The choice of sawdust size was deemed crucial for achieving low $\tan \delta$ values in the mixtures.

Dispersion test results

The dispersion test results can be found in Table 2, and the histogram of agglomerates is presented in Fig. 10. The obtained results in this study indicate that the wood sawdust filler material has a significant effect on the dispersion and aggregate size in natural rubber composites. The filler material does not distribute homogeneously within the natural rubber matrix, as evidenced by the decrease in the dispersion percentage with increasing filler content. Furthermore, there

Table 2 Dispersion test results

Spot	<i>x</i>	<i>y</i>	<i>z</i>	Dispersion (%)	Average agg. size (μm)	Agg. size SD (μm)
Natural Rubber (Ref.)	7.85	9.86	99.36	99.78	5.25	7.99
NR + WS10A	3.89	4.92	90.84	96.79	2.81	10.46
NR + WS10A	3.40	1.33	89.29	96.25	2.54	11.17
NR + WS10A	2.33	1.00	84.29	94.50	2.93	11.39
NR + WS10B	3.39	5.91	89.38	96.28	4.07	11.95
NR + WS20B	3.35	2.92	89.30	96.25	3.84	13.64
NR + WS30B	2.29	1.70	84.08	94.43	4.08	13.74
NR + WS10C	3.99	5.92	91.04	96.88	4.25	12.82
NR + WS20C	3.66	1.77	90.01	96.50	3.54	13.77
NR + WS30C	2.98	1.87	87.73	95.71	3.72	13.57

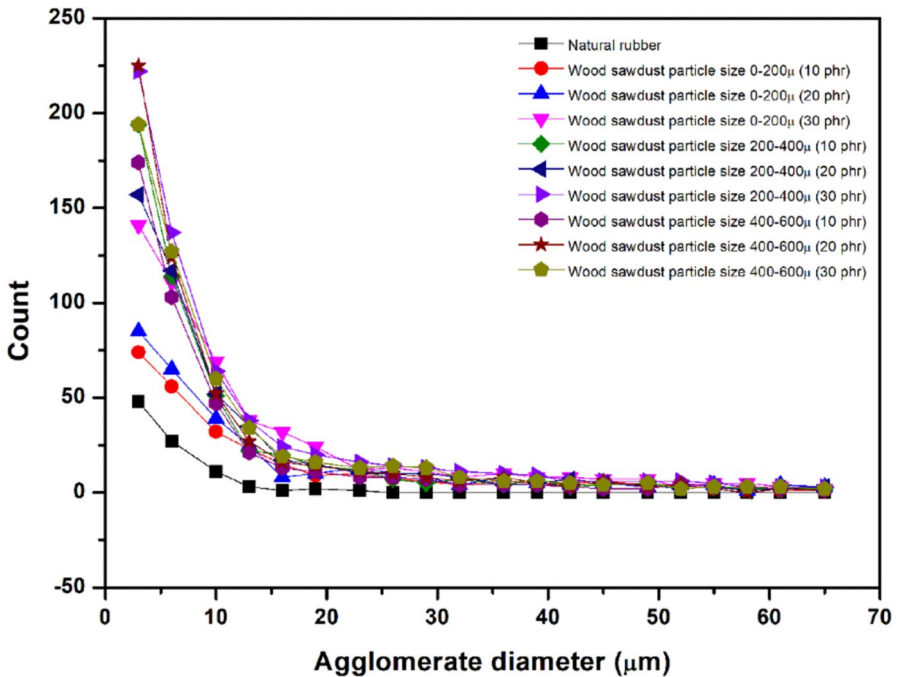


Fig. 10 Histogram of agglomerates

is a correlation between the filler content, particle size, and average aggregate size, where an increase in filler content and particle size generally leads to an increase in the average aggregate size. These findings demonstrate that different filler types and contents can alter the properties of natural rubber composites and influence the material’s homogeneity.

Conclusion

The study's comprehensive analysis and findings lead to the conclusion that integrating wood sawdust, particularly in conjunction with carbon black, presents a promising option as a filler material to enhance both the mechanical and thermomechanical properties of natural rubber composites. The results clearly indicate that introducing wood sawdust as a filler has a substantial impact on several properties of the rubber composites.

1. The optimization of wood sawdust-filled rubber composites was achieved through an increase in filler content and a reduction in particle size, resulting in higher hardness and density.
2. The tensile strength of wood sawdust-filled rubber composites exhibited variations dependent on the filler content. Optimal tensile strength was observed with 10 phr filler content featuring 0–200 μm particle size, but an increase in filler content beyond this level led to a decline in strength. These trends remained consistent across different particle sizes.
3. The incorporation of wood sawdust filler had an adverse impact on tear resistance. To enhance tear resistance, opting for lower filling ratios and smaller particle sizes is recommended. Rebound resilience and permanent deformation were minimally affected by the filler, with a slight increase in rebound resilience observed at higher filling ratios. Reducing permanent deformation can be achieved by maintaining low filling ratios and increasing sawdust size.
4. Accelerated aging tests demonstrated a decrease in tensile strength after air aging, which was mitigated by maintaining low filling ratios, regardless of sawdust size. Elastomer frequency scanning revealed that G^* values were proportional to hardness, while $\tan \delta$ values were inversely proportional to dynamic properties. Higher filling ratios and smaller particle sizes led to increased G^* values, whereas smaller sawdust sizes resulted in lower $\tan \delta$ values. Dispersion tests underscored wood sawdust's significant impact on dispersion and aggregate size, with increased filler content reducing dispersion percentage, indicating a non-homogeneous distribution.

In conclusion, this study offers valuable insights into the potential advantages and constraints associated with integrating wood sawdust as a filler in natural rubber composites. The results provide guidance for optimizing filler content and particle size to enhance specific properties such as hardness, tensile strength, tear resistance, rebound resilience, and dispersion. Further research endeavors can explore additional parameters and conditions to gain a comprehensive understanding of the impact of wood sawdust filler on the performance and diverse applications of natural rubber composites. Based on the findings discussed in the conclusion section, wood sawdust-filled natural rubber composites demonstrate potential for various applications where enhanced mechanical and thermomechanical properties are critical. These composites could be particularly advantageous in the automotive industry, serving in applications such as gaskets,

seals, and vibration dampers, where improved durability and performance are essential. Additionally, the enhanced properties make these composites suitable for construction materials, contributing to their resilience and longevity in demanding environments.

Author contribution KI and KĪ wrote the main manuscript. The experimental design in the article was developed by KI, and all authors contributed to the execution and interpretation of the tests. All authors reviewed the manuscript.

Data Availability The raw data required to reproduce these findings can be obtained from the authors via email.

Declarations

Conflict of interest The authors declare no competing interests.

References

1. Prakash SO, Sahu P, Madhan M, Johnson Santhosh A (2022) A review on natural fibre-reinforced biopolymer composites: properties and applications. *Int J Polym Sci* 2022:1–15
2. Corinaldesi V, Mazzoli A, Siddique R (2016) Characterization of lightweight mortars containing wood processing by-products waste. *Constr Build Mater* 123:281–289
3. Joseph S, Joseph K, Thomas S (2006) Green composites from natural rubber and oil palm fiber: physical and mechanical properties. *Int J Polym Mater Polym Biomater* 55(11):925–945
4. Jacob M, Francis B, Thomas S, Varughese KT (2006) Dynamical mechanical analysis of sisal/oil palm hybrid fiber-reinforced natural rubber composites. *Polym Compos* 27(6):671–680
5. Jacob M, Thomas S, Varughese KT (2004) Mechanical properties of sisal/oil palm hybrid fiber reinforced natural rubber composites. *Compos Sci Technol* 64(7–8):955–965
6. Goettler LA, Shen KS (1983) Short fiber reinforced elastomers. *Rubber Chem Technol* 56(3):619–638
7. Raj RG, Kokta BV, Maldas D, Daneault C (1989) Use of wood fibers in thermoplastics. VII/ The effect of coupling agents in polyethylene–wood fiber composites. *J Appl Polym Sci* 37(4):1089–1103
8. Raj RG, Kokta BV (1991) Reinforcing high density polyethylene with cellulosic fibers. I: the effect of additives on fiber dispersion and mechanical properties. *Polym Eng Sci* 31(18):1358–1362
9. Yam KL, Gogoi BK, Lai CC, Selke SE (1990) Composites from compounding wood fibers with recycled high density polyethylene. *Polym Eng Sci* 30(11):693–699
10. Kadlcak J, Kuritka I, Tunnicliffe LB, Cermak R (2015) Rapid Payne effect test—a novel method for study of strain-softening behavior of rubbers filled with various carbon blacks. *J Appl Polym Sci* 132:41976. <https://doi.org/10.1002/app.41976>
11. Li C, Ding Y, Yang Z, Yuan Z, Ye L (2020) Compressive stress-thermo oxidative ageing behaviour and mechanism of EPDM rubber gaskets for sealing resilience assessment. *Polym Testing* 84:106366. <https://doi.org/10.1016/j.polymertesting.2020.106366>
12. Phuihangpa N et al (2020) IOP Conf Ser: Mater Sci Eng 773:012013
13. Kartal İ, Nayci G, Demirel H (2020) The effect of chestnut wood flour size on the mechanical properties of vinyl ester composites. *Emerging Mat Res* 9(3):960–965
14. İlyas K, Hilal S (2024) Usability of pine sawdust and cotton together as filler in recycled polypropylene composites. *Int J Comput Exp Sci Eng* 10(2):1. <https://doi.org/10.22399/ijcesen.244>
15. Kaewpruk C, Boopasiri S, Poonsawat C, Sae-Oui P, Siri Wong C (2021) Utilization of sawdust and wood ash as a filler in natural rubber composites. *ChemistrySelect* 6:264
16. Ahmad I, Mei TM (2009) Mechanical and morphological studies of rubber wood sawdust-filled UPR composite based on recycled PET. *Polym-Plast Technol Eng* 48(12):1262–1268. <https://doi.org/10.1080/03602550903204105>

17. Shakir MA, Ahmad MI, Yusup Y et al (2023) From waste to wealth: converting rubber wood sawdust into green mycelium-based composite. *Biomass Conv Bioref*. <https://doi.org/10.1007/s13399-023-05113-9>
18. Manaila E, Stelescu MD, Craciun G, Ighigeanu D (2016) Wood sawdust/natural rubber ecocomposites cross-linked by electron beam irradiation. *Materials* 9:503. <https://doi.org/10.3390/ma9070503>
19. Sombatsompop N, Kantala C, Wimolmala E (2006) Wood sawdust fibres as a secondary filler in carbon black filled NR vulcanizates. *Polym Polym Compos* 14(4):331–348
20. Varghese S, Kuriakose B, Thomas S, Koshy TA (1991) Dynamic mechanical properties of short sisal fibre reinforced natural rubber composites. *Indian J Nat Rubb Res* 4(1):55–67
21. Geethamma VG, Joseph R, Thomas S (1995) Short coir fiber-reinforced natural rubber composites: effects of fiber length, orientation, and alkali treatment. *J Appl Polym Sci* 55(4):583–594
22. Chakraborty SK, Setua DK, De SK (1982) Short jute fiber reinforced carboxylated nitrile rubber. *Rubber Chem Technol* 55(5):1286–1307
23. Setua DK, De SK (1983) Short silk fiber reinforced natural rubber composites. *Rubber Chem Technol* 56(4):808–826
24. Arumugam NKTS, Selvy KT, Rao KV, Rajalingam P (1989) Coconut-fiber-reinforced rubber composites. *J Appl Polym Sci* 37(9):2645–2659
25. Ismail H, Shuhelmy S, Edyham MR (2002) The effects of a silane coupling agent on curing characteristics and mechanical properties of bamboo fibre filled natural rubber composites. *Eur Polymer J* 38(1):39–47
26. Iannace S, Ali R, Nicolais L (2001) Effect of processing conditions on dimensions of sisal fibers in thermoplastic biodegradable composites. *J Appl Polym Sci* 79(6):1084–1091
27. Matar M.S. (2022) Investigating the impact and mechanical properties of reclaimed rubber and sawdust composite. Master Thesis, Qatar University, Qatar
28. Kartal İ, Uzunyol CS (2024) Investigation of mechanical properties of walnut shell filled polyester based composites. In: 9th International Biltek congress on current developments in science, technology and social sciences, Hakkari University—Türkiye, Proceedings Book
29. Gao T, Xie R, Zhang L, Gui H, Huang M (2015) Use of rubber process analyzer for characterizing the molecular weight parameters of natural rubber. *Int J Polym Sci*. <https://doi.org/10.1155/2015/517260>
30. Dick JS, Harmon C, Vare A (1999) Quality assurance of natural rubber using the rubber process analyzer. *Polym Testing* 18(5):327–362
31. Liu H, Wang X, Jia D (2020) Recycling of waste rubber powder by mechano-chemical modification. *J Clean Prod* 245:118716
32. Hadi NA, Shuib RK (2019) Effect of plasticizer on microstructure and dynamic mechanical performance of anisotropic magnetorheological elastomers. In: IOP conference series: materials science and engineering, vol 548, No 1, 012007). IOP Publishing
33. Ciullo PA, Hewitt N (1999) *The rubber formulary*, 1st edn. Noyes Publication, New York
34. Ciesielski A (1999) *An introduction to rubber technology*, 1st edn. Smithers Rapra Publications, Shawbury
35. Simpson RB (2002) *Rubber basics*, 1st edn. Smithers Rapra Publications, Shawbury
36. Dick JS (2009) *Rubber technology: compounding and testing for performance*. Hanser, Munich

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