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# Improvement of Mechanical and Viscoelastic Properties of Polypropylene with Wood and Wollastonite Fillers

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# Abstract

In recent years, polypropylene has become one of the most popular thermoplastic polymers due to its excellent properties, chemical resistance, affordability, and easy processing. When reinforced with glass fibers, wood fibers, or mineral fillers, polypropylene composites exhibit enhanced mechanical strength, stiffness, and durability. This makes them suitable for applications where high performance and specific mechanical properties are required, such as in automotive interior parts, building materials, and consumer goods. This research analyzes the effects of wood fibers as an organic filler and wollastonite mineral as an inorganic filler on the mechanical and viscoelastic properties of polypropylene. Various weight ratios of these fillers were added into polypropylene to produce hybrid biocomposites using a laboratory-type highspeed thermokinetic mixer and a heated-cooled hydraulic press. The mechanical properties were determined by tensile and three-point bending tests, and viscoelastic properties were analyzed using dynamic mechanical analysis. The test results indicated that the polypropylene composite sample containing 7% by weight of silane-treated wollastonite and 3% by weight of wood fibers showed the best results among all samples. The storage and loss moduli of the sample are approximately 25% and 22% higher than those of the polypropylene, respectively. Overall, hybrid biocomposites filled with silane-treated wollastonite exhibited enhanced mechanical and viscoelastic properties compared to those filled with untreated wollastonite, as supported by the experimental data.

Keywords: Organic and inorganic fillers, Hybrid composites, Mechanical properties, Polypropylene, Silane-treated wollastonite, Viscoelastic properties

# 1. Introduction

sustainable The increasing demand for and environmentally friendly materials has led to а significant increase in interest in biocomposites, which are composites manufactured from natural fibers and biobased matrices. These materials offer a number of advantages over conventional composites, including a reduced environmental impact, biodegradability and a lower cost [1]. The use of biocomposites is becoming increasingly prevalent across a range of industries, including automotive, construction and packaging. This is due to the favorable mechanical properties exhibited by these materials, which include a high strength-toweight ratio and good thermal stability [2]. Among biocomposites, hybrid composites, which combine two

or more types of fibers or fillers, have attracted considerable attention due to their potential to enhance performance characteristics while maintaining environmental benefits. The selection of appropriate fibers and fillers allows the creation of hybrid composites that can be tailored to achieve a balance between mechanical strength, stiffness, and other desired properties [3, 4]. The utilization of hybrid composites incorporating natural fillers, such as wood flour or wood fibers, in conjunction with mineral fillers, including wollastonite, can present distinctive benefits, including enhanced stiffness, improved dimensional stability and reduced water absorption [5].

Wollastonite, a naturally occurring calcium silicate mineral, has been effectively used as a filler in composite materials due to its ability to improve mechanical properties, thermal stability and fire resistance [6]. When incorporated into hybrid composites, wollastonite can



provide significant improvements in wear resistance and thermal performance, which are crucial for applications in automotive components and construction materials [7]. Similarly, wood fibers, derived from renewable sources, are gaining popularity due to their low density, high availability, and biodegradability, making them ideal for eco-friendly composite applications [8-10]. The combination of wood fibers and wollastonite in hybrid biocomposites represents a promising avenue for the development of advanced materials with enhanced mechanical, thermal, and environmental properties. These hybrid composites offer a more sustainable alternative to traditional synthetic composites and align with the increasing regulatory and consumer demands for greener and more sustainable products [11, 12]. The objective of this paper is to examine the potential of hybrid biocomposites reinforced with wollastonite and wood fibers, with a focus on their properties, processing methods, and potential applications in various industries.

There is a growing need for materials that offer both high performance and sustainability. Polypropylene (PP) is a popular thermoplastic due to its chemical resistance, low cost, and ease of processing. When reinforced with fillers like glass fibers, wood fibers, or minerals, PP composites become stronger and more durable, making them ideal for use in automotive parts, construction, and consumer products. This study explores the effects of adding wood fibers and wollastonite, a mineral filler, to PP to create hybrid biocomposites. By examining how different amounts of these fillers affect the mechanical and viscoelastic properties of PP, this research aims to find new ways to improve material performance while using more sustainable resources.

This work observes the impact of incorporating wood fibers and wollastonite, a mineral filler, into PP to create hybrid biocomposites. The objective of this research is to identify new methods of enhancing the mechanical and viscoelastic properties of PP while utilizing more sustainable resources. To achieve this, an examination was conducted on the effects of varying quantities of fillers on the properties of PP.

# 2. Materials and Methods 2.1. Materials

The PP-copolymer (LG Chem M1500) used in this work that has a density of 0.9 g/cm3. Untreated wollastonite (Tremin 939-300) and silane-treated wollastonite (Tremin 939-300 AST), both with needle-like morphology, a density of 2.85 g/cm<sup>3</sup>, and a Mohs hardness of 4.5, were obtained from Kaolin Minerals, Türkiye. The woods were supplied from the products left as pruning waste from a cherry tree plant field in Konya, Türkiye. To make wood fibers suitable for composite production, branches were broken into small pieces then ground with a laboratory-type grinder. Then, wood fibers were passed through sieves. Particles, under 100 micrometer sizes, were used to produce composites. The production of polymer composites was carried out to a high-speed thermokinetic mixer and a heated-cooled hydraulic press. The nomenclature of the produced samples is provided in Table 1 for reference.

**Table 1.** The nomenclature of the composite samples

Name	Rates (wt. %)
PP	100% Polypropylene
3UTW-	3% Untreated wollastonite + 7% Wood fiber
7WF	+ 90% PP
7UTW-	7% Untreated wollastonite + 3% Wood fiber
3WF	+ 90% PP
3STW-	3% Silane-treated wollastonite + 7% Wood
7WF	fiber + 90% PP
7STW- 3WF	7% Silane-treated wollastonite + 3% Wood fiber + 90% PP

#### 2.2. Methods

The mechanical properties-including tensile strength, tensile modulus, flexural strength, and flexural modulus-of the samples were measured using a tensile testing machine (Shimadzu AGS-X, 5 kN, Japan). Tensile tests were conducted following ASTM D638-14, "Standard Test Method for Tensile Properties of Plastics," with a cross-head speed of 50 mm/min. Flexural tests adhered to ASTM D790-17, "Standard Test Methods for Flexural Properties of Unreinforced and Reinforced Plastics and Electrical Insulating Materials," with a cross-head speed of 1 mm/min. Each test was repeated at least five times for each material type to ensure accuracy and reliability. Dynamic mechanical properties, including storage modulus and loss modulus, were measured using a dynamic mechanical analyzer (TA Instruments Inc., USA). The analyses were conducted with a single-point holder over a temperature range of 35-130°C, with a heating rate set to 3°C/min.

# 3. Results and Discussion 3.1. Tensile Test

The tensile test results illustrated in Figure 1. The tensile strength of PP was found to be  $23.16 \pm 0.18$  MPa, as given in Figure 1.a. Among the composites, the 7STW-3WF sample exhibited a tensile strength of  $23.06 \pm 0.67$ MPa, which is in close proximity to that of PP. These findings suggest that the incorporation of 7% silanetreated wollastonite maintains the tensile strength of PP, with the addition of wood fiber having a negligible effect. contrast, composites comprising untreated In wollastonite (3UTW-7WF and 7UTW-3WF) exhibited slightly reduced tensile strengths of  $21.02 \pm 0.21$  MPa and  $21.89 \pm 0.07$  MPa, respectively. This indicates that silane treatment enhances tensile strength by improving interfacial adhesion between the wollastonite and the PP matrix [6,13].





Figure 1. a) Tensile strength and b) Tensile modulus

The incorporation of fillers resulted in a notable enhancement in the tensile modulus of the composites, as seen in Figure 1.b, in comparison to PP, which exhibited a tensile modulus of  $912.07 \pm 76.97$  MPa. The highest tensile modulus was observed in the 7STW-3WF sample, with a value of  $1221.66 \pm 15.78$  MPa, indicating that the addition of 7% silane-treated wollastonite effectively increases the stiffness of the composite. This improvement in stiffness is likely due to enhanced bonding between the treated wollastonite and the PP matrix, which restricts the mobility of the polymer chains [14]. The tensile modulus values for the composites with untreated wollastonite (3UTW-7WF and 7UTW-3WF) were also higher (1142.85  $\pm$  36.2 MPa and 1131.60  $\pm$ 14.26 MPa, respectively) than those of the pure PP, although these values were slightly lower than those of the silane-treated samples. This indicates that silane treatment has a beneficial effect on tensile properties.

# 3.2. Flexural Test

The flexural test results demonstrated in Figure 2. The flexural strength of PP was determined to be  $32.63 \pm 0.7$ 

MPa, as shown in Figure 2.a. The incorporation of fillers led to an enhancement in flexural strength, with the 7STW-3WF composite exhibiting the highest value of  $39.25 \pm 0.62$  MPa. This increase can be attributed to the reinforcing effect of both the silane-treated wollastonite and wood fibers, which enhance the load distribution across the composite matrix [6]. The 3UTW-7WF and 7UTW-3WF samples also exhibited enhanced flexural strengths (38.23  $\pm$  0.7 MPa and 37.46  $\pm$  1.59 MPa, respectively) in comparison to PP. However, these increments were less pronounced than those observed in the silane-treated samples. This highlights the efficacy of surface treatment in enhancing interfacial bonding and optimizing composite performance.



Figure 2. (a) Flexural strength and (b) Flexural modulus

The flexural modulus, which reflects the material's resistance to bending, was significantly enhanced in all composite samples compared to PP, which had a modulus of 966.13  $\pm$  53.69 MPa (Figure 2.b). The 7STW-3WF sample exhibited the highest flexural modulus at 1571.87  $\pm$  37.43 MPa, followed closely by the 3STW-7WF sample at 1551.27  $\pm$  37.81 MPa. Composites with untreated wollastonite (3UTW-7WF



and 7UTW-3WF) also demonstrated considerable improvements in flexural modulus ( $1488.89 \pm 88.95$  MPa and  $1466.8 \pm 42.53$  MPa, respectively), though these values were slightly lower than those of the silane-treated samples. The increased flexural moduli in the silanetreated composites are likely due to better compatibility and stress transfer between the fillers and the PP matrix, which reduces deformation under bending loads [12].

The research indicates that incorporating silane-treated wollastonite into PP composites significantly improves their tensile and flexural properties compared to using untreated wollastonite. The 7STW-3WF composite, in particular, achieved the best overall mechanical performance, indicating an optimal balance of filler content for reinforcing PP while maintaining cost-effectiveness and environmental sustainability. These findings are consistent with previous research demonstrating that surface treatment of fillers enhances their interfacial bonding with the polymer matrix, leading to improved mechanical properties [3].

#### 3.3 Dynamic Mechanical Analysis (DMA)

Figure 3 illustrates the variation of both the storage modulus and loss modulus with temperature for PP and various hybrid biocomposites reinforced with untreated and silane-treated wollastonite and wood fibers. The storage modulus, representing the material's stiffness, indicates its ability to store energy elastically under deformation, providing insight into the viscoelastic behavior of the composites under different temperatures—a key factor in their real-world performance. In contrast, the loss modulus measures the energy dissipated as heat during deformation, reflecting the material's damping characteristics and viscous response to stress. This parameter is essential for understanding the energy absorption and internal friction behavior of the composites under dynamic loading conditions.

Figure 3.a illustrates that as the temperature increases, the storage modulus of PP decreases significantly, starting from around 1000 MPa at 30°C and declining steadily to about 200 MPa at 160°C, which reflects the typical thermal softening behavior of thermoplastic polymers as they become less stiff and more compliant due to increased molecular mobility [15]. The 3UTW-7WF composite exhibits a higher initial storage modulus than PP, starting at around 1200 MPa at 30°C, indicating that the addition of untreated wollastonite and wood fiber enhances the stiffness of the composite at lower temperatures due to the reinforcing effects of the fillers [16]. However, as the temperature increases, the modulus decreases at a rate similar to that of PP, showing a notable reduction in stiffness at higher temperatures. In comparison, the 7UTW-3WF sample starts with a slightly higher initial storage modulus of around 1300 MPa at 30°C, suggesting that a greater content of wollastonite contributes more effectively to stiffness. Although its modulus also decreases with increasing temperature, this composite retains higher stiffness than both the 3UTW-7WF and PP throughout the temperature range, emphasizing the role of wollastonite in maintaining the material's stiffness.



Figure 3. (a) Storage modulus and (b) Loss modulus

The 3STW-7WF composite shows a higher initial storage modulus than its untreated counterparts, starting at approximately 1350 MPa at 30°C. This increase is attributed to the silane treatment, which enhances interfacial adhesion between the wollastonite particles and the PP matrix, leading to improved stress transfer and increased stiffness [17]. As the temperature rises, the modulus decreases, but it remains consistently higher than that of the untreated composites, indicating superior thermal stability. The 7STW-3WF sample shows the highest initial storage modulus of all samples, beginning around 1400 MPa at 30°C, and demonstrates the slowest rate of decline in modulus with increasing temperature. maintaining a significantly higher modulus than all other samples even at 160°C. These findings clearly highlight the combined effect of higher wollastonite content and silane treatment in enhancing both the stiffness and thermal stability of the composite material.



The loss modulus of PP, given in Figure 3.b, shows a characteristic peak around 90°C, corresponding to the glass transition temperature ( $T_g$ ) of the polymer, where there is a marked increase in molecular mobility leading to maximum energy dissipation. Beyond the  $T_g$ , the loss modulus decreases steadily as the polymer transitions into a rubbery state, becoming more flexible and less resistant to deformation [15].

The 3UTW-7WF composite exhibits a higher loss modulus than PP, with a peak value slightly shifted to around 95°C, suggesting that the addition of untreated fillers enhances the energy dissipation capacity of the composite, likely due to increased internal friction between the matrix and the fillers. However, the peak height indicates that while the composite dissipates more energy, it also experiences increased internal friction, which could negatively affect its dynamic mechanical performance at elevated temperatures [16]. Similarly, the 7UTW-3WF composite shows an even higher loss modulus compared to PP, with a peak similar to that of 3UTW-7WF but slightly greater, implying that a higher wollastonite content contributes to increased damping properties. This may result from the rigid nature of wollastonite particles, which provide more resistance to deformation and, consequently, greater energy dissipation.

The 3STW-7WF sample exhibits a peak loss modulus that is higher than those of both the untreated composites and PP, with the peak occurring around 100°C, slightly above that of the untreated counterparts. This shift and increase in peak height indicate improved interfacial bonding between the matrix and the treated filler due to the silane treatment, which enhances stress distribution and increases frictional damping within the composite [17]. The 7STW-3WF sample shows the highest peak loss modulus, also occurring around 100°C. This elevated peak and shift further highlight the effectiveness of silane treatment in enhancing the composite's damping properties. The higher wollastonite content in this composite results in a greater interfacial area and more pronounced interaction with the polymer matrix, leading to increased energy dissipation and improved thermal stability [18].

The results show that the addition of both untreated and silane-treated wollastonite and wood fibers enhances the stiffness and damping properties of PP composites, with silane-treated wollastonite showing superior performance in both aspects. The 7STW-3WF composite exhibited the highest storage modulus across all temperatures tested, making it the most suitable candidate for applications requiring high stiffness and thermal stability. This improved performance can be attributed to better interfacial bonding provided by the silane treatment, which facilitates effective load transfer between the filler and the polymer matrix [18]. Additionally, the higher wollastonite content (7%)

consistently results in greater stiffness compared to a higher wood fiber content (7%) in both treated and untreated groups, suggesting that wollastonite is a more effective reinforcement than wood fiber [19]. Similarly, silane-treated fillers enhance the loss modulus of the composites, with the 7STW-3WF composite showing the highest loss modulus and the most significant shift in T<sub>g</sub>, indicating superior damping capacity and energy dissipation. This improved damping behavior is likely due to better adhesion between the filler and the matrix, which facilitates more efficient stress transfer and energy dissipation [20]. The findings suggest that wollastonite's rigid structure and surface characteristics make it more effective in improving both the stiffness and damping properties of PP composites compared to wood fiber.

# 4. Conclusion

The study demonstrates that the incorporation of both untreated and silane-treated wollastonite and wood fibers enhances the mechanical and viscoelastic properties of PP composites. The incorporation of these fillers has been observed to enhance the tensile and flexural modulus, thereby indicating an increase in stiffness and load-bearing capacity when compared to PP. Among the reults, composites with silane-treated wollastonite (3STW-7WF and 7STW-3WF) exhibited superior mechanical performance, with the highest tensile and flexural moduli. This indicates that surface treatment facilitates improved interfacial bonding between the filler and the matrix, enabling effective stress transfer and enhanced resulting in mechanical properties. Furthermore, dynamic mechanical analysis confirms these findings, demonstrating that silane-treated wollastonite composites exhibit elevated storage and loss moduli across the investigated temperature range. This suggests enhanced energy storage and dissipation capabilities due to augmented matrix-filler interaction. Furthermore, the shift in the glass transition temperature (T<sub>g</sub>) to higher values in treated composites indicates enhanced thermal stability and reduced mobility of polymer chains, which can be attributed to the formation of strong interfacial bonds between the fillers and the matrix. In conclusion, the results indicate that silane treatment of wollastonite is an effective strategy for enhancing the mechanical and dynamic properties of PP composites. Composites with higher wollastonite content, particularly the 7STW-3WF formulation, exhibit the most promising performance, combining high stiffness, strength, and damping properties. This makes them suitable for applications requiring higher mechanical and viscoelastic properties, such as automotive components, construction materials, and consumer products. Further studies could concentrate on optimizing filler content and investigating additional surface treatments with a view to enhancing the performance of these hybrid biocomposites.



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### **Author's Contributions**

**Mustafa Öncül:** Performed the experiment and result analysis, drafted and wrote the manuscript.

**Kutlay Sever:** supervised the experiment's progress, result interpretation and helped in manuscript preparation.

# Ethics

There are no ethical issues after the publication of this manuscript.

#### References

[1]. Ashori A. Wood–plastic composites as promising greencomposites for automotive industries! Bioresour Technol. 2008; 99(11):4661–7.

[2]. Faruk O, Bledzki AK, Fink HP, Sain M. Biocomposites reinforced with natural fibers: 2000–2010. Top Issue Polym Biomater. 2012 Nov 1; 37(11):1552–96.

[3]. Koronis G, Silva A, Fontul M. Green composites: A review of adequate materials for automotive applications. Compos Part B Eng. 2013; 44(1):120–7.

**[4].** Savran M, Öncül M, Yılmaz M, Aydın L, Sever K. Modeling and optimization of dynamic-mechanical properties of hybrid polymer composites by multiple nonlinear neuro-regression method. Sigma J Eng Nat Sci. 2023; 41(6):1243–54.

[5]. Mohan Sharma A. Mechanical Behaviour, Water Absorption and Morphology of Wheat Straw, Talc, Mica and Wollastonite filled Polypropylene Composites [Internet] [Master's Thesis]. University of Waterloo; 2012 [cited 2024 Sep 10]. Available from: https://uwspace.uwaterloo.ca/handle/10012/6772

**[6].** Chan JX, Wong JF, Hassan A, Mohamad Z, Othman N. Mechanical properties of wollastonite reinforced thermoplastic composites: A review. Polym Compos. 2020 Feb; 41(2):395–429.

**[7].** Karagöz İ, Büyükkaya K, Demirer H, Mudu M, Kartal İ. Mechanical and thermal characterization of elastomer modified polypropylene hybrid composites reinforced with hazelnut shell and wollastonite fillers. J Appl Polym Sci. 2024 Aug 10; 141(30):e55710.

**[8].** Atagür M, Kaya N, Uysalman T, Durmuşkahya C, Sarikanat M, Sever K, et al. A detailed characterization of sandalwood-filled high-density polyethylene composites. J Thermoplast Compos Mater. 2020 Jul 31; 089270572093915.

**[9].** Savran M, Yılmaz M, Öncül M, Sever K. Manufacturing and Modeling of Polypropylene-based Hybrid Composites by Using Multiple-Nonlinear Regression Analysis. Sci Res Commun. 2022 Jan 30; 2(1):1–15.

**[10].** Sever K, Yılmaz M. Evaluation of Mechanical and Thermal Properties of Artichoke Filled Polypropylene Composites: Influence of Wollastonite Hybridization. Emerg Mater Res. 2020 Jun 1;9(2):1–6.

[11]. Geissdoerfer M, Savaget P, Bocken NM, Hultink EJ. The Circular Economy–A new sustainability paradigm? J Clean Prod. 2017; 143:757–68.

[12]. Mohanty AK, Misra M, Drzal LT. Natural fibers, biopolymers, and biocomposites. CRC press; 2005.

[13]. Chen M, Wan C, Shou W, Zhang Y, Zhang Y, Zhang J. Effects of interfacial adhesion on properties of polypropylene/Wollastonite composites. J Appl Polym Sci. 2008 Feb 5; 107(3):1718–23.

**[14].** Wong JF, Chan JX, Hassan AB, Mohamad ZB, Othman NB. Thermal and flammability properties of wollastonite-filled thermoplastic composites: a review. J Mater Sci. 2021 May; 56(15):8911–50.

[15]. Andrady AL. Science and technology of polymer nanofibers [Internet]. John Wiley & Sons; 2008 [cited 2024 Sep 10]. Available from:

https://books.google.com/books?hl=en&lr=&id=KOdoYIuFHzAC&oi =fnd&pg=PR7&dq=Science+and+Technology+of+Polymer+Nanofib ers&ots=ue1XtfEz31&sig=HkcLBWLddnWjU5ezGCQL4INGYYY

[16]. Joseph P, Joseph K, Thomas S, Pillai C, Prasad V, Groeninckx G, et al. The thermal and crystallisation studies of short sisal fibre reinforced polypropylene composites. Compos Part Appl Sci Manuf. 2003; 34(3):253–66.

[17]. Luyt AS, Dramićanin MD, Antić Ž, Djoković V. Morphology, mechanical and thermal properties of composites of polypropylene and nanostructured wollastonite filler. Polym Test. 2009; 28(3):348–56.

**[18].** Fu SY, Lauke B, Mäder E, Yue CY, Hu X. Tensile properties of short-glass-fiber-and short-carbon-fiber-reinforced polypropylene composites. Compos Part Appl Sci Manuf. 2000; 31(10):1117–25.

[19]. Lee SY, Kang IA, Doh GH, Yoon HG, Park BD, Qinglin Wu. Thermal and Mechanical Properties of Wood Flour/Talc-filled Polylactic Acid Composites: Effect of Filler Content and Coupling Treatment. J Thermoplast Compos Mater. 2008 May; 21(3):209–23.

[20]. Malkapuram R, Kumar V, Yuvraj Singh Negi. Recent Development in Natural Fiber Reinforced Polypropylene Composites. J Reinf Plast Compos. 2009 May; 28(10):1169–89.