Effect of Particle Size and Loading of Cherry Tree Branch Fillers on the Mechanical and Viscoelastic Properties of Polypropylene Composites

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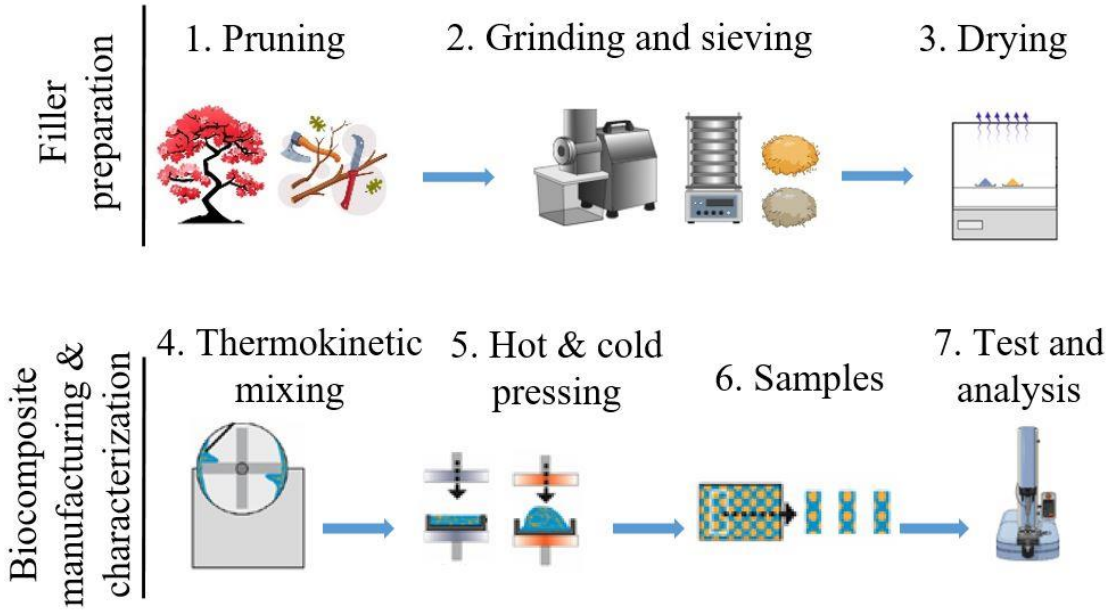
GRAPHICAL ABSTRACT



Effect of Particle Size and Loading of Cherry Tree Branch Fillers on the Mechanical and Viscoelastic Properties of Polypropylene Composites

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Lignocellulosic fillers derived from pruned cherry tree branches were studied relative to the mechanical and viscoelastic properties of polypropylene (PP) composites. Tree branches were collected from the orchard after pruning and the wood and bark parts were separated from each other. Both materials were processed into particles of different sizes (below 100 µm and between 100 and 250 µm) and filled into PP at different weight percentages (5%, 10%, 15%, and 20%). The mechanical performances of the biocomposites were evaluated through tensile tests, while their viscoelastic behavior was analyzed using dynamic mechanical analysis (DMA). Results revealed a decline in tensile strength with increasing filler content, which was attributed to poor interfacial adhesion between the PP matrix and fillers. However, tensile modulus increased with increasing filler content, with the highest values were observed at 20% filler loadings. The DMA showed enhanced storage and loss moduli, indicating improved stiffness and energy dissipation. Scanning electron microscopy (SEM) confirmed the presence of voids and filler agglomeration, further explaining the mechanical property reductions. These results demonstrate the potential of cherry tree pruning waste as a bio-filler for sustainable biocomposites with improved stiffness.

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Keywords: Lignocellulosic fillers; Cherry tree pruning waste; Mechanical properties; Polypropylene; Viscoelasticity; Wood; Bark

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INTRODUCTION

In recent years, natural fiber-reinforced polymer composites have gained significant attention due to their sustainability, eco-friendliness, and cost-effectiveness (Elfaleh *et al*. 2023; Palanisamy *et al*. 2024). Among various natural fibers, wood-based materials, such as wood fibers, offer a promising alternative to synthetic fibers in polymer composites (Skosana *et al*. 2024). These bio-fillers not only enhance the mechanical properties of polymers but also contribute to the reduction of plastic waste by partially replacing petroleum-based polymers. Studies have demonstrated that incorporating wood fibers into polymer matrices such as polypropylene (PP) can enhance specific mechanical properties, such as stiffness and impact resistance, while also modifying thermal, dynamic, and viscoelastic behaviors, making these biocomposites suitable for diverse industrial applications (Sanjay *et al*. 2016).

Recent studies have explored various wood fibers and fillers to enhance the properties of PP composites. Reixach *et al.* (2015) investigated orange wood fibers as reinforcement for PP, finding that while the fibers did not alter transition temperatures,

they increased polymer crystallinity by 7% at 50% fiber content. Similarly, Atagur *et al.* (2020) demonstrated that incorporating up to 20% of *Ceratonia siliqua* powder significantly improved tensile and flexural strength by 32% and 23%, respectively, while reducing the coefficient of thermal expansion. Hernández-Jiménez *et al.* (2022) examined white oak wood flour, noting increases in Young's and flexural moduli with particle incorporation, though reduction in elongation and strength were observed. Andrzejewski *et al.* (2024) evaluated wood-PP composites under multiple processing cycles, observing a decline in viscosity and mechanical properties due to PP matrix degradation.

The use of pruning wastes in polymer composites has emerged as an innovative approach to both waste management and sustainable material development. Agricultural and forestry by-products, such as pruned branches, have traditionally been considered as low-value waste; they are often discarded or burned, thereby contributing to environmental pollution (Reixach *et al.* 2015). However, recent studies highlight their potential as reinforcements/fillers in polymer composites, particularly in enhancing mechanical and thermal properties (Maraveas 2020; Öncül *et al.* 2024). Incorporating pruning wastes, such as branches from cherry trees, into polymer matrices not only helps in reducing the dependency on virgin wood and synthetic fibers but it also provides a sustainable alternative for producing high-performance materials (Cabrera 2021). The environmental benefits of utilizing pruning residues extend beyond waste reduction; they also decrease the overall carbon footprint of composite production by replacing energy-intensive synthetic materials (Gupta *et al*. 2022). According to the Turkish Statistical Institute (2023), the total number of fruit-bearing trees in Turkey is approximately 900 million, of which 2.5% are cherry trees. In the calculation of pruning waste, the pruning coefficient, which varies according to the type of fruit tree, was reported as 5.90 for cherry trees (Bilandzija *et al.* 2012). According to these assumptions, the total amount of cherry tree pruning waste was estimated as 131 thousand tons/year for cherry trees.

This study addresses both environmental and material challenges by investigating the potential of using wood and bark derived from pruning waste of cherry trees, at two different particle sizes and different filling ratios, as a sustainable reinforcement in PP matrix. To our knowledge, no previous research has explored the incorporation of cherry tree branch particles into PP composites. This study examined the mechanical and viscoelastic properties of these biocomposites, focusing on key performance metrics, such as tensile strength, stiffness, and dynamic-mechanical behavior. The results will contribute to the development of sustainable, high-performance materials for industries such as automotive, construction, and packaging.

EXPERIMENTAL

Materials

The matrix material used in this study is commercial polypropylene (PP, LG M1500, South Korea), a widely utilized thermoplastic in polymer composite applications due to its favorable mechanical properties and chemical resistance. It has a melt flow index of 16 g/10 min (230 °C, 2.16 kg) and a density of 0.9 g/cm^3 as physical characteristics. Cherry tree (*Prunus avium* L.) pruning branches, were used as a lignocellulosic filler in this study. These agricultural wastes were collected from an orchard at an altitude of about 1600 m in the Taurus Mountains in the Mediterranean region of Turkey (Konya, Turkey). The age of the trees in the garden is in the range of 5 to 10 years old.

Methods

The wood (W) and bark (B) components of the pruned cherry branches were first separated to facilitate their processing. Following, the W and B samples were processed separately using a laboratory grinder (Mertest LB160, Mertler Machine, Eskişehir, Turkey). After grinding, the particles were sieved using a sieve shaker (Retsch RS200, Retsch GmbH, Haan, Germany), with sieving performed at both 100 µm and 250 µm. This process enabled the classification of the fillers into two distinct particle size ranges: those below 100 µm (W1 and B1) and those between 100 and 250 µm (W2 and B2). Biocomposite samples were produced using a high-speed thermokinetic mixer and a laboratory type hot-cold hydraulic press (Gülnar Machine, Gülnar, Kayseri, Turkey) (Öncül 2023). The nomenclature of the matrix material, fillers, and biocomposites used in this study is presented in Table 1.

Table 1. Nomenclature of Materials

Abbreviations	Samples
W1	Wood fillers below 100 µm particle size
W2	Wood fillers between 100-250 µm particle size
B1	Bark fillers below 100 µm particle size
B2	Bark fillers between 100-250 µm particle size
PP	Neat polypropylene
5W1-10W1- 15W1-20W1	5-10-15-20 wt% wood-filled PP below 100 µm particle size
5W2-10W2-15W2-20W2	5-10-15-20 wt% wood-filled PP between 100-250 µm particle size
5B1-10B1-15B1-20B1	5-10-15-20 wt% bark-filled PP below 100 µm particle size
5B2-10B2-15B2-20B2	5-10-15-20 wt% bark-filled PP between 100-250 µm particle size

A universal testing machine (Shimadzu AG-IC, Shimadzu Corporation, Kyoto, Japan) with a 5 kN load cell was used to determine the mechanical properties of the samples. Tensile tests were conducted in accordance with the ASTM D638-14 (2014), which specifies procedures for testing the tensile properties of plastics, with a crosshead speed of 50 mm/min. Tensile tests were repeated at least five times per sample to ensure statistical reliability and minimize experimental error. Viscoelastic properties of samples were determined using a dynamic mechanical analyzer (DMA Q800, TA instruments, New Castle, DE, USA). Analyses were performed with a single cantilever clamp at a temperature range of 30 to 140 °C and heating rate of 3 °C/min. The morphological properties of the fractured surfaces of the samples were detected with a scanning electron microscope (SEM) (Carl Zeiss 300VP, Zeiss, Oberkochen, Germany) at an accelerating voltage of 5 kV. Before observation, the samples were uniformly coated with gold using a sputter coating device (Quorum Q150 Res, Quorum, East Sussex, UK) to prevent charging under the electron beam.

RESULTS AND DISCUSSION

Tensile Test

The tensile strength and tensile modulus values of PP and biocomposites are illustrated in Figs. 1. As shown in Figs. 1a and 1b, the tensile strength of PP shows a notable decline with the incorporation of fillers. Specifically, the tensile strength of PP decreased from 22.2 to 16.2 MPa for sample 20B2. This reduction in tensile strength can be attributed to the interaction between the fillers and the matrix. First, it appears that the W and B particles are not sufficiently wetted by the PP matrix, which may hinder effective stress transfer (Shumigin *et al*. 2011; Pickering *et al*. 2016). Additionally, the fillers were not uniformly dispersed within the polymer matrix, and the interfacial adhesion between the non-polar PP and the polar lignocellulosic particles was weakened. This weak interfacial bonding becomes more pronounced as the particle loading increases, as a higher filler content results in a greater interfacial surface area, exacerbating the poor adhesion between the matrix and the fillers, further compromising the tensile strength of the composite (Rosa *et al*. 2009; Kılınç *et al*. 2018; Atagur *et al*. 2020). Furthermore, the presence of non-wetted wood clusters may contribute to failure due to insufficient bonding between the fillers and the matrix (Hubbe *et al*. 2023).

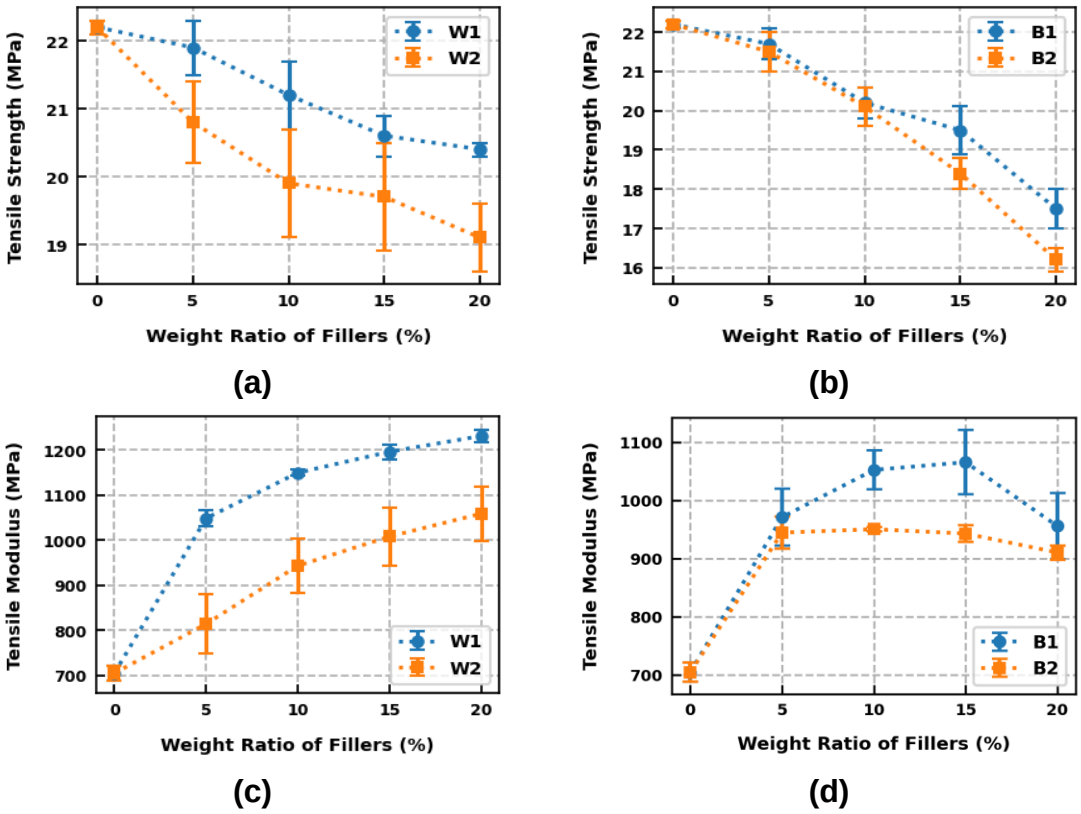

Fig. 1. Tensile test results of PP and biocomposites

Conversely, changes in the tensile modulus of PP and biocomposites are presented in Figs. 1(c-d). The tensile modulus of PP was measured at 704 MPa, and an increase in tensile modulus was observed with the addition of 5% filler. This trend continued consistently with higher filler contents, reaching a peak at 20% filler loading. The highest

tensile modulus was observed in the 20W1 sample, with a value of 1230 MPa, representing a 75% increase compared to PP. This important enhancement in tensile modulus is due to the lignocellulosic fillers having a much higher modulus than the polymer matrix, thereby stiffening the biocomposite as the filler content increases. Similarly, previous studies revealed a decline in tensile strength and an increase in tensile modulus with the addition of wood filler (Pérez *et al.* 2012; Ndiaye *et al.* 2013; Soccalingame *et al.* 2015).

Dynamic Mechanical Analysis

The variations in the storage modulus of the materials as a function of temperature are presented in Figs. 2(a–b). The results indicate that the storage modulus of the biocomposites was consistently higher than that of PP across the entire temperature range. This suggests that the addition of fillers improved the stiffness of the PP. The reduction in modulus with increasing temperature is likely due to the initiation of the relaxation process and softening of the polymer matrix, which becomes more prominent as the material heats up (Sarikanat *et al*. 2014). The rise in storage modulus with the incorporation of fillers is attributed to the mechanical constraints introduced by the embedded particles within the polymer matrix, which restricts chain mobility and enhances rigidity. A comparable decline in the storage modulus was also observed in the study conducted by Reixach *et al.* (2015).

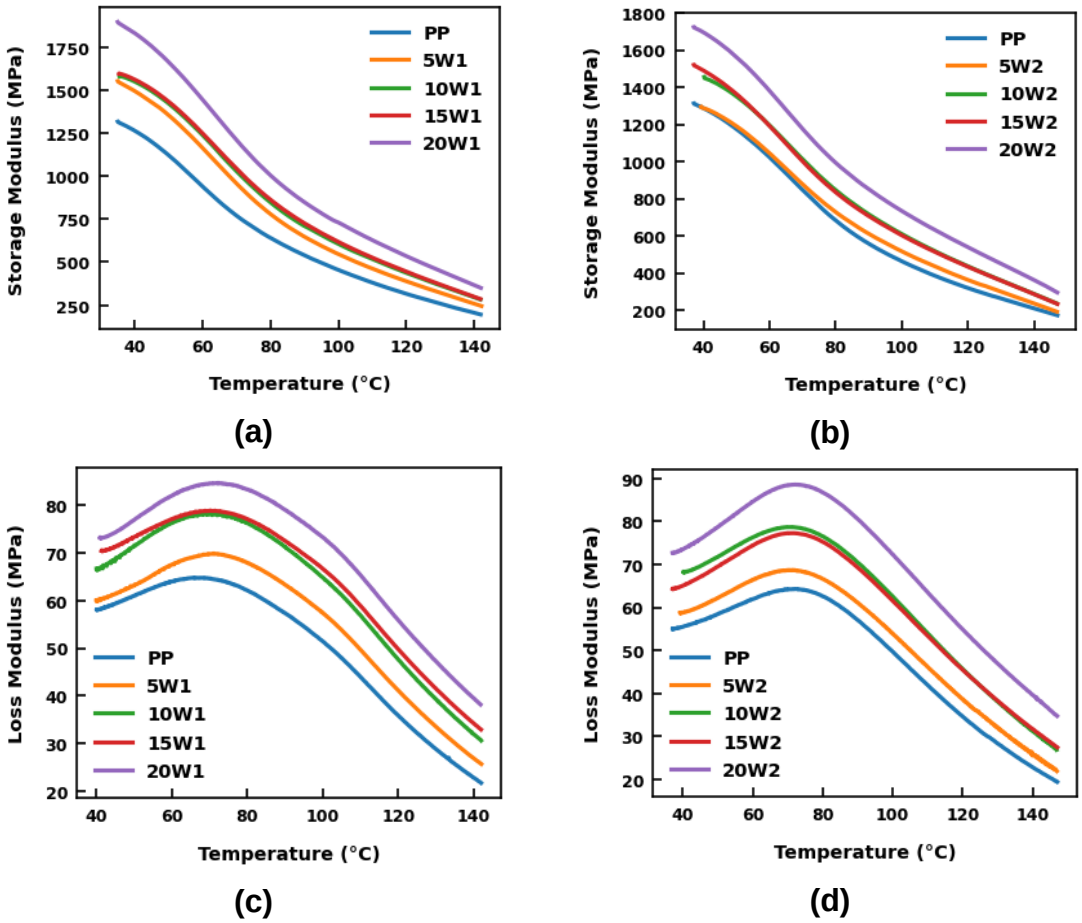

Fig. 2. Dynamic mechanical analysis results of PP and biocomposites

Similarly, the changes in loss modulus as a function of temperature are shown in Figs. 2(c–d). The biocomposites exhibit higher loss modulus values than PP, with the highest loss modulus observed in samples filled with 20 wt% of filler, irrespective of particle size. This increase in loss modulus indicates greater energy dissipation in the biocomposites, which may be associated with enhanced internal friction and reduced energy absorption efficiency as the filler content increases (Seki *et al*. 2013; Nagarajan *et al*. 2016). Reixach *et al.* (2015) observed a similar trend in the loss modulus throughout the entire temperature range in the study.

Scanning Electron Microscopic Analysis

The lignocellulosic fillers are clearly observable within the PP matrix upon examining the fracture surfaces of the biocomposites, as shown in Figs. 3(a–d). The presence of numerous small pores around the filler particles suggests poor matrix-filler interfacial adhesion. These pores indicate weak compatibility between the matrix and the fillers, which greatly impacts the mechanical performance of the biocomposites. As the filler content increases, the dispersion of the particles within the matrix becomes less homogeneous, further aggravating the poor interfacial bonding. This is consistent with findings from other studies, which highlight that the increased filler content can lead to agglomeration, reducing the effectiveness of stress transfer across the matrix-filler interface (Babaei *et al*. 2014).

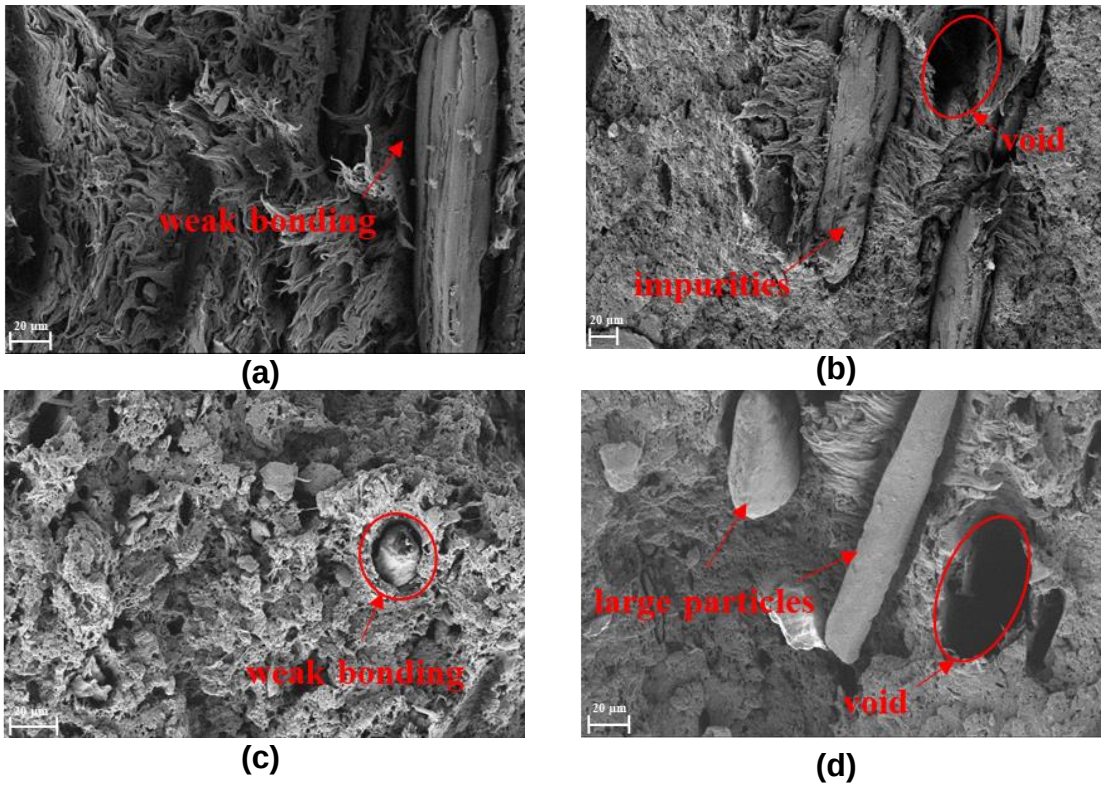

Fig. 3. SEM images of the fracture surfaces: a) 10W1, b) 10W2, c) 10B1, and d) 10B2

In Fig. 3d, it is evident that larger filler particles began to dominate the fracture surface as the filler content increased. This accumulation of larger particles created stress concentration points, which weakened the composite's ability to resist tensile forces.

Consequently, the tensile strength of the biocomposite decreased with higher filler loadings. Similar trends have been observed in other studies, where excessive filler addition has led to poor dispersion and weakened mechanical properties due to ineffective stress transfer and the formation of voids at the interface (Xie *et al*. 2010; Andrzejewski *et al*. 2024). This highlights the importance of optimizing filler content and particle dispersion to balance mechanical strength and filler reinforcement in polymer composites.

In the previous study, Öncül *et al.* (2024) observed that the cellulose content of wood was approximately 11% higher than that of bark, while the hemicellulose and lignin content of wood were 22% and 23% lower than that of bark, respectively. This finding aligns with the results of other similar studies that have also reported significant differences between wood and bark in terms of physical, chemical, and mechanical properties (Chow *et al.* 2008; Ruiz *et al.* 2015).

CONCLUSIONS

1. This study examined the influence of lignocellulosic fillers derived from pruned cherry tree branches on the mechanical and viscoelastic properties of polypropylene (PP) composites.
2. The particle size comparison of wood fillers demonstrated that the composites prepared with smaller cherry branch particles (W1) exhibited higher tensile strength due to enhanced dispersion and stress transfer within the PP. The incorporation of larger particles in W2 resulted in a notable decline in strength, predominantly due to the formation of voids and a concomitant reduction in adhesion. A similar pattern was observed for the particle size comparison of bark fillers.
3. The results of the tensile tests and dynamic mechanical analysis (DMA) consistently demonstrated that wood-filled composites outperformed bark-filled composites. The superior structure and composition of wood fibers facilitated better dispersion, resulting in better properties compared to bark fillers.
4. It can be stated that lignocellulosic fillers derived from pruned cherry tree branches, particularly those of under 100 µm particle size, demonstrate potential for use in the reinforcement/filler of PP.

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