UTILIZATION OF WASTE CELLULOSIC FIBERS AND FINES TOWARD HIGH-PERFORMANCE POLYPROPYLENE COMPOSITES: EFFECTS OF COMPATIBILIZER AND COMPOUNDING SPEED

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Abstract

This study investigated the feasibility of transforming waste cellulosic fiber and fines (WCFF) from the pulp and paper industry into value-added products in the form of high-performance polypropylene (PP) composites. Polypropylene and cellulose-based reinforcement composite systems suffer from incompatibility and uniform mixing challenges. Polypropylene is a non-polar hydrophobic polymer, while cellulose is a polar, hydrophilic material due to the abundance of hydroxyl groups. To enhance the compatibility and performance of PP/20%WCFF composites, this research studied the effects of compatibilizer content and compounding screw speed on various properties of the composites. PP was compounded with 20 wt.% WCFF in the presence of varying percentages of maleic-anhydride grafted PP (MA-g-PP: 0-5 wt.%). The compounding was conducted in a twin-screw extruder at three screw speeds of 300, 600, and 900 rpm. The composites' rheological, mechanical, and thermal behaviors were thoroughly evaluated. The results showed that incorporating MA-g-PP into the PP composite at an optimal amount of 3 wt.% increased the tensile strength and tensile modulus while reducing the viscosity. Crystallinity was also slightly increased with the addition of MA-g-PP, probably because of the increased crystal nucleation sites due to a better WCFF dispersion state. The screw speed also revealed significant effects on the mechanical behavior, viscosity, and crystallinity. Overall, lower screw speed favored more enhanced mechanical properties. The optimal performance was achieved with 3 wt.% MA-g-PP and a screw speed of 300 rpm, which improved fiber dispersion and minimized fiber breakage and thermal degradation, resulting in a tensile strength of 41.29 MPa and modulus of 3,207 MPa—an improvement of 27.2 and 37.7%, respectively, compared to those of pure unfilled PP. This work suggests that optimized formulation and compounding processes can deliver significantly enhanced performance in PP by incorporating waste cellulosic materials to develop more sustainable composites.

Keywords: Cellulose, Waste, Compatibility, Recycling, Composite, Processing, Sustainability

1 Introduction

Using waste cellulosic fibers in polypropylene (PP) offers several potential benefits, including increased sustainability by utilizing a renewable resource, reduced environmental impact due to waste reduction, improved mechanical properties like strength and stiffness when adequately incorporated, potentially increased biodegradability, lower cost compared to pristine cellulosic materials and synthetic fibers, and the ability to tailor material properties depending on the application; all while contributing to a more circular economy by repurposing waste materials [1,2]. The primary challenge however in creating PP and cellulose-based composites is the inherent incompatibility between the hydrophilic nature of cellulose fibers and the hydrophobic PP matrix, leading to poor interfacial adhesion and uneven dispersion of cellulose fibers, significantly impacting the mechanical properties of the composite material. This often requires additional processing steps such as surface modification of cellulose fibers to enhance compatibility and achieve optimal performance [2,3]. Extensive research has been conducted on surface modification and utilization of coupling agents to enhance the hydrophilicity of polypropylene. Maurya et al.[4] conducted a comprehensive review of natural fiber-reinforced PP hybrid composites. The study focused on various types of fibers, such as sisal, flax, kenaf, hemp, and coir. The review highlighted the poor interfacial adhesion of natural fibers with the PP matrix and proposed additional treatments and process optimizations to improve mechanical interlocking.

Compatibilization is a promising and scalable route that has been utilized to enhance the interaction between surfaces of natural fiber and polymeric matrices [4]. According to Lu et al. [5], a material that reduces the interfacial energy between two separate phases or improves the dispersion of fibers within the matrix is considered a compatibilizer. Compatibilizers such as monofunctional isocyanates and maleic anhydride grafted PP (MA-g-PP) are commonly used in various applications. Maleic anhydride grafted PP facilitates esterification and hydrogen bonds between the -OH group of the fiber and the -MA groups of MA-g-PP. There is also evidence of chain entanglement between MA-g-PP and PP during compatibilization, which occurs when MA-g-PP is added to the natural fiber and polymer composite system [4]. Borja et al. [6] reported the mechanical and thermal properties of PP reinforced with pulp and Lyocell fibers. They compared the content of the fiber and the addition of MA-g-PP. The MA-g-PP reduced the crystallinity in the case of pulp, but it induced crystallization when lyocell was used. However, the fibers used were not waste materials, rather chemically extracted cellulose fiber. Hernández-Díaz et al. [7] worked on the impact strength and water uptake of composites made of virgin PP reinforced with fibers from newspapers; however, no compatibilizer was used, so limited miscibility was achieved. Also, Benhamadouche et al. [8] developed a composite using jute recovered from packaging bags and PP reclaimed from scraps from manufacturing PP yarn with miscibility challenges. In another study by Franco-Marquès et al. [9], the influence of the intrinsic parameters of 6% of maleated PP on the mechanical properties of PP/fiber composites were examined. Highly functionalized maleated PP coupling led to important improvements in mechanical strength and water resistance due to the formation of stronger interfaces. However, the report did not evaluate MA-g-PP variation. All other studies, e.g., [9–11], used treated and well controlled wood-based pulp (like pure cellulose). The motivation for this work was to assess the performance of PP composites reinforced with as-received cellulose waste material with minimal treatments as an economically viable recycling strategy.

The process conditions of compounded composites have been reported to affect their output properties significantly. Berzin et al [12] highlighted the importance of optimizing the compounding speed to limit fiber degradation while improving properties of the lignocellulosic fiber-based composite. Specifically, the screw speed of the compounding machine has a significant effect on the fiber distribution and fiber integrity, which affect the functional properties of the composite [12]. Bouaziz et al. [13] reported that increasing the screw speed during the processing of a PP composite generally leads to better dispersion of the reinforcing fillers within the polymer matrix, resulting in improved mechanical properties like tensile strength, impact strength, and elongation at break, but can also potentially cause increased polymer degradation and severe fiber aspect ratio reduction if the speed is too high. There are limited studies on the effect of screw speed on PP-reinforced cellulosic-based fiber.

This study explores the effect of compatibilizer and compounding speed on PP reinforced with WCFF. The percentage of MA-g-PP and the screw speed were varied to understand their impact on various properties and determine their optimum values for 20% WCFF fiber addition. The PP pellets were mixed with WCFF, compounded, and pelletized. Compression molding and microinjection molding were then used to prepare samples for tensile and rheological characterizations. Morphological, thermogravimetry and wettability analysis were also conducted and the relationships between these characteristics and MA-g-PP content and screw speed were discussed. The optimal processing conditions for enhanced mechanical strength were also identified.

2 Materials and Method

2.1 Materials

The waste cellulosic fiber and fine (WCFF) was received in pellet form (about 6 mm in diameter and 20 mm in length), supplied by American Wood Fibers, Columbia, MD, USA. It had an average density of 1.3 g/cm^3 and a 4-5 wt.% moisture content. The experiments utilized PP Impact Copolymer (PPC), provided by AMCO Polymers, Orlando, Florida, USA (Grade: PPC O50-30 NA) as the baseline virgin PP. The tensile strength (yield), tensile elongation (yield), and flexural modulus (1% secant) of the PP were 33.43 MPa, 7.0%, and 1,689 MPa, respectively. The resin had a density of 0.9 g/cm^3 With a load of 2.16 kg, its melt flow rate (MFR) was 5.0 g/10min at 230 °C. Maleic anhydride grafted polypropylene (MA-g-PP) (Grade: Orevac-CA100) was supplied by SK-functional polymer Courbevoie, France, was utilized as a coupling agent to enhance the compatibility between the WCFF and the matrix (PP). At 230 °C with a load of 2.16 Kg, the MFR of the MA-g-PP was 10 g/10min and it had a density of 0.91 g/cm^3.

2.2 Composite preparation

The WCFF pellets were first dissociated into loose microfiber and fines by a simple 60 sec. blending process. It was then dried using a VWR scientific 1350 FM forced air oven, Radnor, PA, at 80 °C for 24 hours to reduce the moisture content to less than 1 wt.%. The PP was also dried at 80 °C and compounded with WCFF using a twin-screw extruder (Model: Leistritz twin-screw extruder, ZSE 18HP) with an 18 mm screw diameter and a 40 L/D ratio.

Table 1 shows the experimental design used to study the effects of MA-g-PP content and compounding screw speed on the performance of PP/20%WCFF composites. MA-g-PP concentrations of 0, 3, and 5 wt.% were examined with PP/20%WCFF to find a proper compatibilizer ratio. The screw speed was also varied over 300, 600, and 900 rpm to discern the impact of shear on the compounding. The mixed PP and MA-g-PP pellets, as well as WCFF feedstock, were fed into the extruder using Brabender FW12-0 feeder and THAYER solid flow control, respectively. A 50 g/min throughput was consistently maintained. The barrel temperature for zone 1 (next to the feed throat) was maintained at 170 °C while the remaining zones 2-8 were maintained at 180 °C. The melt pressure measured before the die exit was between 1.10 and 1.17 MPa; and the extruder level was consistently 31-34% of the maximum torque. The extrudates were chilled in an ice-water bath after extrusion and pelletized using a Bay Plastics Machinery pelletizer BT25. The pelletized compounds were then dried in an air oven at 80°C for 24 h to remove any moisture. The compounds were compression molded into flat sheets with a thickness of 0.62 mm, length of 126.38 mm, and width of 111.14 mm. The molding process was carried out at a temperature of 180 °C and a pressure of 10.3 MPa using a CARVER Inc. 4394.4PL3003 compression molder. After keeping the compression condition constant for 5 min to stabilize the sheet, it was cooled to 20 °C in 5 min using a water-based cooling system. The samples were then die cut for tensile testing. The as-compounded pellets were used for TGA and DSC tests. A plunger micro-injection molding machine (Xplore IM 12) set at 200 °C was used to make disc-shaped rheology samples of 25 mm in diameter and 1.5 mm in thickness.

Table 1 Experimental design to study the effects of MA-g-PP content and compounding screw speed on the performance of PP/20%WCFF composites

Sample	PP (wt.%)	MA-g-PP (wt.%)	Cellulose (WCFF) (wt.%)	Screw Speed (rpm)
PP	100	0	-	300
PP/MA3/R300	97	3	-	300
PPF/MA0/R300	80	0	20	300
PPF/MA3/R300	77	3	20	300
PPF/MA5/R300	75	5	20	300
PPF/MA3/R600	77	3	20	600
PPF/MA3/R900	77	3	20	900

Note: PPF- Polypropylene with WCFF, MA -maleic anhydride with the number representing its wt.%, R- compounding speed with the number representing rpm.

2.3 Characterization

The melt flow characteristics were evaluated using a TA Instruments ARES G2 rheometer, which had a 25 mm stainless steel parallel plate geometry with a 1.5 mm gap. At a fixed strain of 5%, measurements were made within the linear viscoelastic zone. At temperatures of 170 and 230 °C, frequency sweeps ranging from 100 to 0.1 rad/s were carried out. The rheology results were used to infer the level of interaction between the fibers and the matrix and the homogeneity of the fiber distribution. The calculated linear viscoelastic functions of complex viscosity, storage modulus (G'), and loss modulus (G") were plotted for discussion.

The tensile properties of the molded samples were obtained using an Instron 5966 with a 10 kN load cell using the ASTM D638 standard (type V) with a modified sample thickness of 0.62 mm. A 10 mm/min displacement rate was used for all the samples. Each sample was evaluated on five specimens, and the mean and standard deviation of the tensile strength, Young's modulus, and elongation at break are reported. With a 5 kV acceleration voltage, a JEOL scanning electron microscope (SEM) (JSM 6390) was used to examine the surface morphology of cryo-fractured composite samples. Cryo-fractured samples were coated with a thin layer of gold using Vacuum Desk IV sputtering equipment for five minutes before SEM analysis.

Thermal characterization from the DSC instrument (METTLER DSC 3+, C151224649) was reported, including crystallinity, melting point, and crystallization temperature. The ASTM D3418-15 standard was followed to investigate the heating and cooling behavior. All composites and the neat PP matrix (5-6 mg) were put in aluminum pans and heated to 250 °C. A Mettler TGA 2 instrument (type CIS 1224647) was employed to evaluate the samples' thermal stability. The samples were weighed to a target range of 8–8.5 mg to guarantee accurate and consistent findings. Next, the samples were heated steadily at 20 °C per minute under a nitrogen environment, from 50 °C to 600 °C. The density of a composite provides information about the material's compounding ratio, weight, and overall quality. The density measurement was conducted using Mettler-Toledo GmbH Im Langacher 44 8606 Greifensee, Switzerland. The water absorption test on the composites was also performed, following ASTM D570. The samples were dried for 6 hours at 60 °C, air-cooled in a desiccator, and then weighed. They were then submerged in 23 °C water for 24 hours, removed, patted dry with a lint-free cloth, and weighed again.

3 Results and Discussion

3.1 Rheological behavior

Rheological analysis is a critical tool for evaluating dispersion and flow properties in fiber-reinforced composites. In this study, rheology offered clues to the effect of compatibilizer content and compounding speed on the viscoelastic behavior of PP composite [14]. Figure 1 presents the composites' viscoelastic properties, with the top row showing variations in MA-g-PP content (300 rpm) and the bottom row showing variations in screw speed (3 wt.% MA-g-PP). The storage modulus (G'), loss modulus (G"), and complex viscosity (η*) as a function of frequency are given in Figure 1 a&d, b&e, and c&f, respectively, measured at two temperatures of 170 and 230 ˚C. The change in MA-g-PP content from 0 to 3 wt.% caused somewhat significant decreases in G', G", and η* throughout the frequency range and at both temperatures; however, the 5 wt.% MA-g-PP composites had slightly higher values than the un-compatibilized samples. The decrease in viscoelastic properties with an increase of MA-g-PP from 0 to 3 wt.% could be attributed to the lubricating effect of the compatibilizer, which had a lower molecular weight [15]. On the other hand, the increased viscoelastic properties upon further increase of MA-g-PP content to 5 wt.% could be related to the presence of excessive functional groups, creating more grafting points between the components, leading to a reduced PP chain mobility and thus a higher overall viscosity and elasticity.

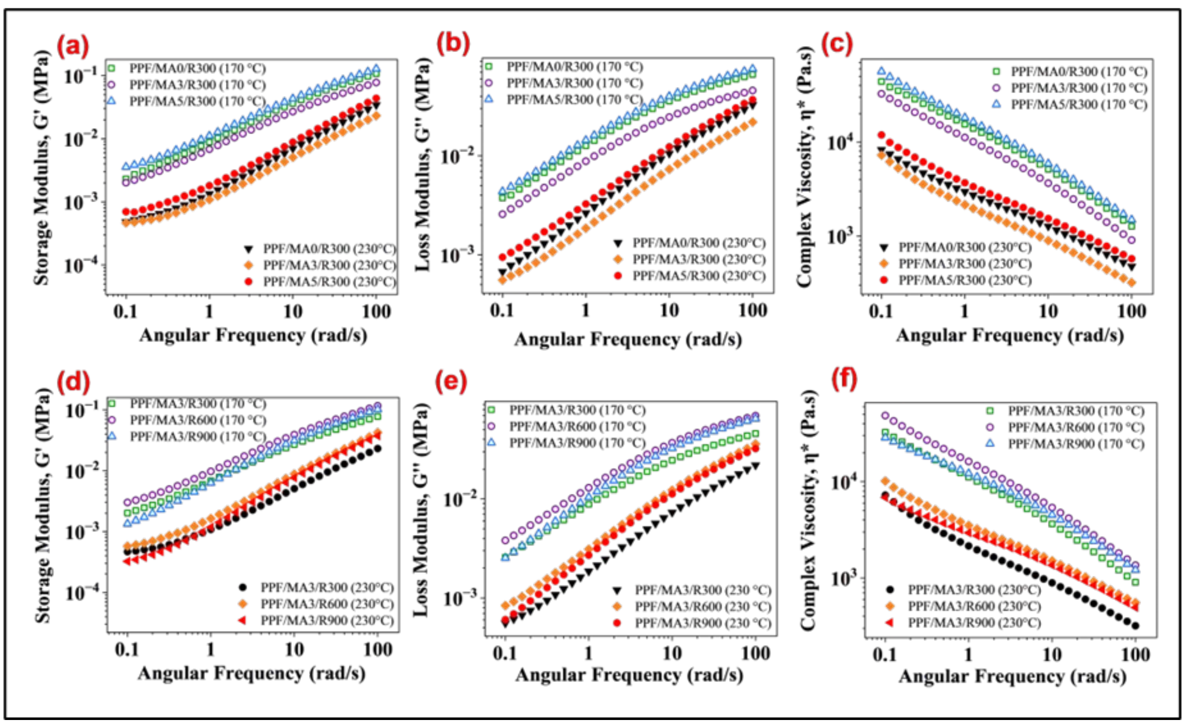

Figure 1 Storage modulus (a), loss modulus (b), and complex viscosity (c) as a function of frequency showing the effect of MA-g-PP content at 170and 230 ˚C; Storage modulus (d), loss modulus (e), and complex viscosity (f) as a function of frequency showing the effect of screw speed at 170 and 230 ˚C.

An increase in the compounding rotational speed also affected the composites' viscoelastic properties as shown in Figure 1d-f. The G', G" and η* values, at 170 and 230 ˚C, increased from 300 to 600 rpm and decreased at 900 rpm with the curves overlapping at some frequencies. The rheology curves at 300 and 600 rpm are quite parallel to each other, indicating that the shear thinning effect is similar, while 900 appears to exhibit a reduced shear thinning effect, which could be related to the degradation of PP or WCFF. Furthermore, between 300 and 600 rpm, improved viscoelastic properties are observed due to the enhanced dispersion of WCFF, stronger interactions between polypropylene and WCF, and better fiber wetting. However, at 900 rpm, excessive shearing has resulted in severe fiber breakage and potential oxidative or thermal degradation, reducing the fiber's aspect ratio and disrupting fiber networks. Consequently, at lower frequencies, the viscoelastic properties tend to align more closely with those observed at 300 rpm, particularly evident in complex viscosity measurements. At higher frequencies, the shortened fibers align more easily with the melt flow, exhibiting a weakened network effect and thus causing the behavior to resemble that at 600 rpm. Overall, the shape of the curve shifts when compared to the similarities observed at 300 and 600 rpm. Previous studies corroborate the results obtained in this study. Increasing the compounding screw speed enhances the viscoelastic properties of a PP fiber composite by promoting better fiber dispersion through increased shear forces which break down fiber agglomerates, leading to a more uniform and consistent material with improved mechanical properties. However, higher screw speeds can also lead to severe fiber breakage and potential fiber and polymer degradation, which could negatively impact the composite's percolated network of solid reinforcement [12,16] reducing mechanical performance as discussed in the next section. Overall, the rheology results reveal the opposing influences of dispersion and interfacial grafting vs fiber breakage and fiber/polymer degradation when extruding these composites at different compatibilizer loadings and screw speeds. The relatively subtle effects suggest that milder compounding and moderate levels of compatibilizer should be sufficient for producing uniformly blended composites.

3.2 Tensile properties

Figure 2a shows the effect of MA-g-PP on the tensile properties. Composite PPF/MA3/R300 containing 3 wt.% MA-g-PP exhibited the best tensile strength (41.29 MPa) and modulus (3,207 MPa) among all three cases. Composite PPF/MA0/R300 and PPF/MA5/R300 demonstrated a tensile strength of 36.59 and 37.61 MPa, respectively. In addition, they recorded tensile modulus of 2,957 and 3,024 MPa, respectively. There was a 24.24% increase in the tensile strength of PPF/MA3/R300 compared to PPF/MA0/R300. Analysis of variance revealed a p-value of 0.00001 < 0.05, which confirms a statistically significant difference between them at a confidence level of 95%. It is also noted that all the composite materials exhibited greater strength and modulus values compared to those of unfilled PP. Elongation at break values exhibited an inverse relationship with the compatibilizer as they decreased up to 3% and then slightly increased at 5 wt.% MA-g-PP. The notable enhancement in the tensile strength and modulus of composite PPF/MA3/R300 can be attributed to the exceptional interfacial interaction between the WCFF and MA group of the coupling agent MA-g-PP. With the addition of 3% MA-g-PP to the blend, the resulting material shows enhanced mechanical strength and reduced melt viscosity (Figure 1c), making it easier to process and mold. As explained in [17,18], due to the reaction between MA-g-PP and cellulose, there is an efficient load transfer from the polymeric matrix to the reinforcing fiber. Nevertheless, when the MA-g-PP content was increased to 5 wt.%, there was a reduction in both tensile strength and modulus, compared to those of PPF/MA3/R300 composite. This drop could be attributed to the inherently lower tensile strength and modulus of the MA-g-PP, compared to the pristine PP [19,20].

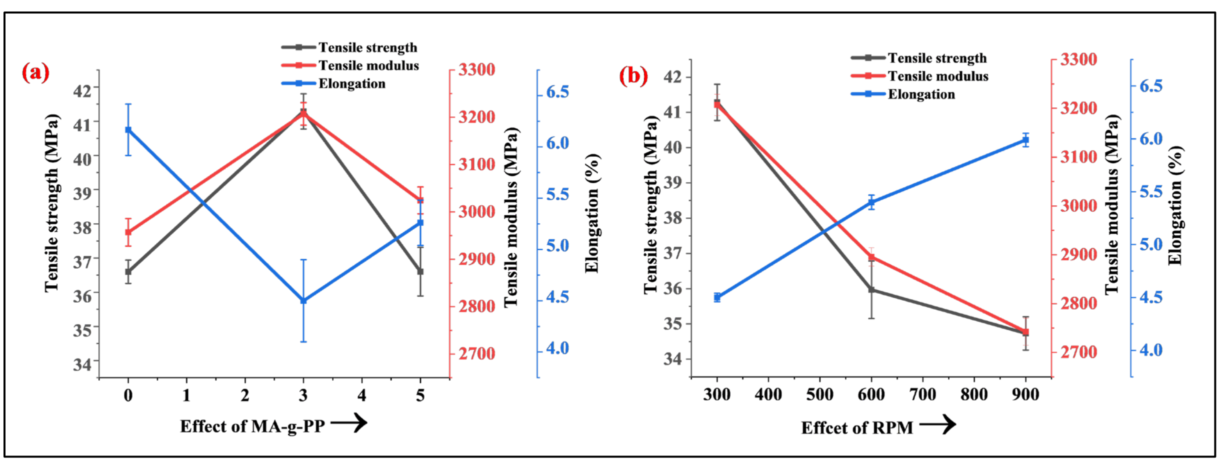

Figure 2 (a) The effect of MA-g-PP content and (b) the effect of screw speed on the tensile strength, modulus, and elongation at break of PP/20%WCFF composites.

Figure 2b shows the effect of screw speed on the tensile properties of the composites. The samples compounded with 300 rpm displayed the maximum tensile strength of 41.28 MPa, whereas those of 900 rpm demonstrated the lowest tensile strength of 34.73 MPa. The tensile modulus was measured to be 3,207 and 2,742 MPa for 300 and 900 rpm cases, respectively, showing a declining trend in their tensile modulus. The 600-rpm case exhibited intermediate values of strength and modulus. Elongation at break values exhibited an inverse relationship with the compounding speed.

Figure 3 shows the SEM micrographs of various composites. The addition of 3 and 5 wt.% MA-g-PP to the composite system led to a better dispersion and distribution of WCFF. The red circled area in Figure 3a (no MA-g-PP) indicates fiber agglomeration, while the red arrows in Figure 3b-c indicate individual fibers (with MA-g-PP), reflecting the effect of MA-g-PP addition. Figure 3d-f also shows the effect of compounding speed on the composite morphology, which is complimented with the optical images, taken from the surface of molded samples, shown in Figure 4a-c. Overall, not a significant difference was observed in the fracture surface of the composites made using various screw speeds. In all, a relatively well distributed WCFF was observed with no significant sign of agglomeration. However, as the screw speed was increased from 300 to 900 rpm, the degree of the material discoloration increased (Figure 4). The intensity of this discoloration is likely to stem from excessive thermal exposure due to higher shear rates at higher speeds.

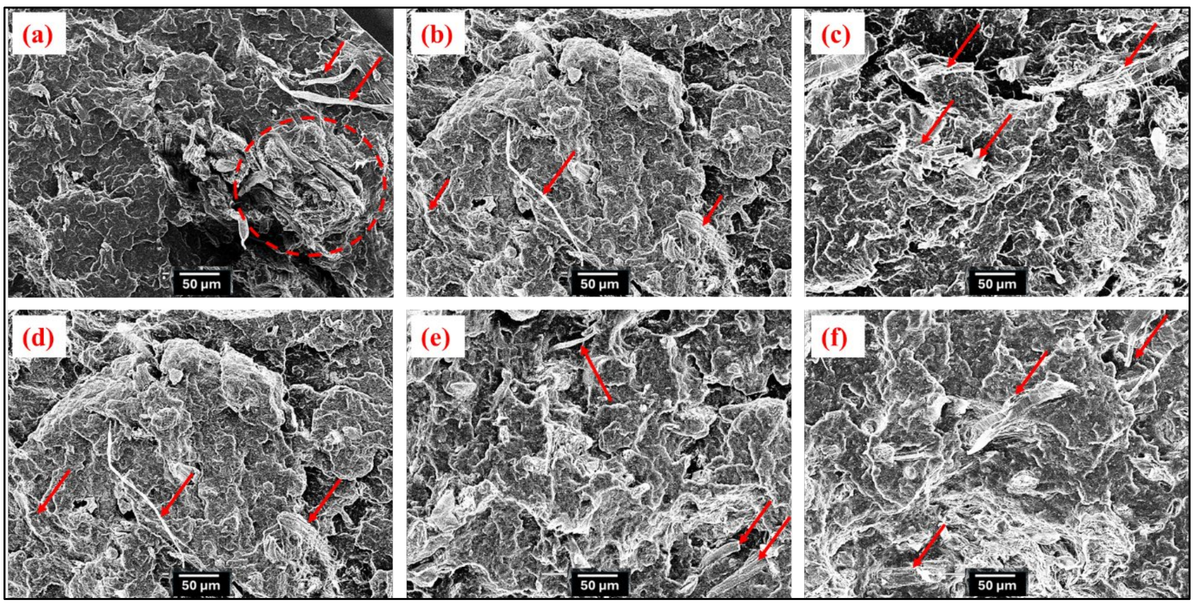

Figure 3 SEM micrographs revealing the composites morphology: The effect of MA-g-PP content: (a) PPF/MA0/R300, (b) PPF/MA3/R300, and (c) PPF/MA5/R300. The effect of screw speed: (d) PPF/MA3/R300, (e) PPF/MA3/R600, and (f) PPF/MA3/R900. The red circle indicates WCFF agglomerate and red arrows indicate individual fibers.

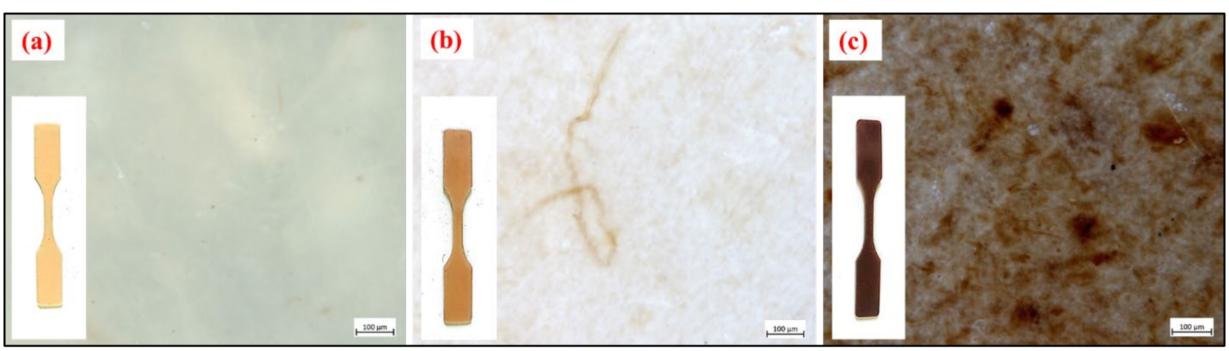

Figure 4 Effect of screw speed on the surface color of the composites: (a) PPF/MA3/R300, (b) PPF/MA3/R600, and (c) PPF/MA3/R900.

A higher screw speed creates greater shear stresses within the melt, which helps to break down agglomerates and distribute filler particles more evenly throughout the polymer matrix, leading to a finer and more homogeneous morphology [21,22]. However, excessively high screw speed results in lower tensile strength and modulus in fiber-reinforced composites, due to increased fiber breakage, which compromises the fiber integrity and reduces the reinforcing effect of the fibers within the polymer matrix. Essentially, higher screw speeds lead to shorter, more fragmented fibers, resulting in a weaker composite material [23]. Furthermore, cellulose and polymer degradation may also occur due to the generation of local excessive viscous heat at high screw speeds. The increased shear stress caused by the rapid rotation leads to significant heat build-up within the material, potentially causing the polymer chains to break down and degrade, essentially "burning" the polymer through excessive mechanical stress [24]. Polymer and fiber degradation can compromise the strength and modulus [25]. These findings show the importance of optimizing the speed to achieve the desired strength and fiber integrity within the polymer composite [20,21].

3.3 Thermogravimetric analysis

Figure 5 shows the thermogravimetry analysis (TGA) and derivative thermogravimetry (DTG) results for PP, WCFF, and PP/20%WCFF composites at various MA-g-PP contents and screw speeds. The WCFF TGA curve dropped slightly initially, probably because of the moisture content. As expected, the TGA and DTG of WCFF and PP show a single degradation curve with the onset temperatures at 195.6 and 377.0 °C and degradation peaks at 356.5 and 462.4 °C, respectively. Overall, the TGA results showed no traceable effect of MA-g-PP on the decomposition behavior of PP/20%WCFF composites. The composites with 0, 3, and 5 wt.% MA-g-PP showed a two-step weight loss with the first onset temperature at 227.2, 227.4 and 227.8 °C and the second onset at 364.8, 364.6, and 364.8 °C, respectively. Similarly, the DTG results for 0, 3, and 5% MA-g-PP showed the first peak at 353.5, 354.0, and 354.8 °C and the second peak at 463.0, 463.3, and 464.0 °C respectively.

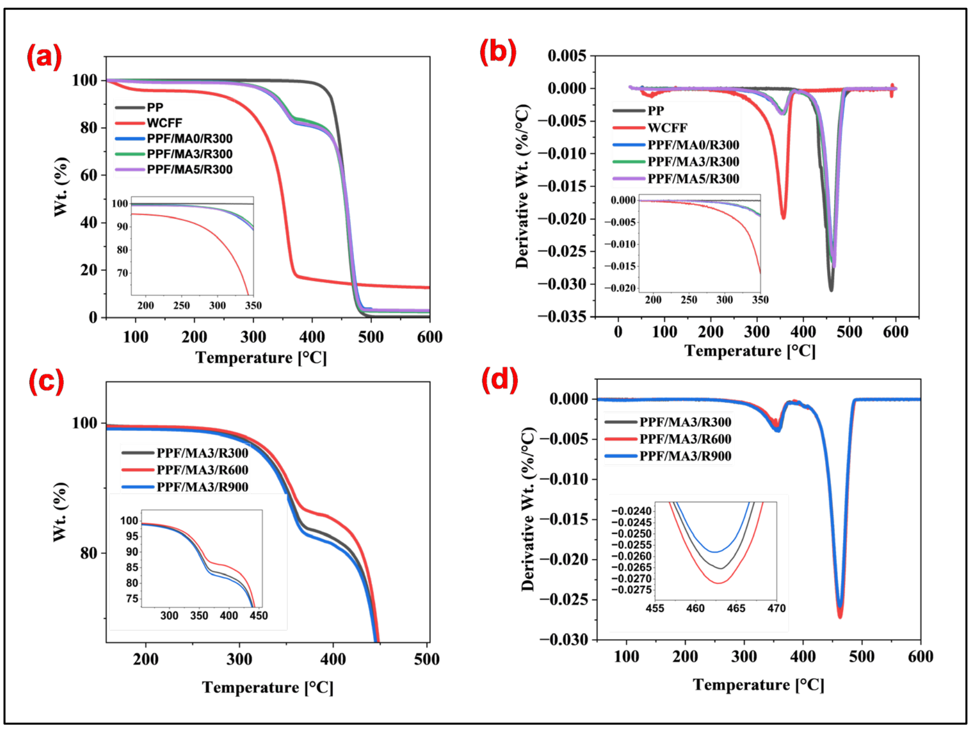

Figure 5 (a) TGA and (b) DTG of PP/20%WCFF with various MA-g-PP content. (c) TGA and (d) DTG of PP/20%WCFF composites with various screw speed.

Overall, the first and second degradation peaks of the composites were close to those of WCFF and PP, respectively. This was expected, knowing that the fiber and the matrix are immiscible and kept as two separate components of the composite. However, the first onset temperature of the composites was about 30 °C higher than that of WCFF itself. This could be an indication of a more stable WCFF in the composite due to the interactions between the PP chains and cellulose at the presence of MA-g-PP [26,27]. Moreover, as seen in Figure 5, the change in MA-g-PP content and screw speed appeared to have an insignificant effect on the thermal decomposition behavior of the PP/WCFF composites. The peak decomposition temperatures and the residues remained largely unaffected by these factors. The measured residues of all the composites were also similar and agreed well with the estimated residue based on the rule of mixture, assuming 20 wt.% WCFF and 80 wt.% PP. This suggests that the variation in the tensile strength and modulus as a function of MA-g-PP content and screw speed were primarily governed by the inherent mechanical properties of MA-g-PP, WCFF, MA-g-PP/WCFF interactions, and fiber size retention after compounding [28].

3.4 Differential scanning calorimetry

Figure 6a-c shows the effect of MA-g-PP on the thermal behavior of PP/20%WCFF composites. The composites with 0, 3, and 5 wt.% MA-g-PP showed crystallization temperatures (T_c) values of 121.7, 121.1, and 121.1 °C, respectively, indicating an insignificant effect of MA-g-PP. However, all these composites reported T_c values lower than that of the virgin PP with a T_c value of 126.7 °C. A similar result was reported for the sunflower hull sanding dust-reinforced composites fabricated by Sui et al. [29], where fillers hindered the mobilization of the PP macromolecular chain and delayed the macromolecular segment from achieving ordered alignment of the crystal structure.

The composites with 0, 3 and 5 wt.% MA-g-PP exhibited 38.8, 46.8, and 46.5 % crystallinity, respectively. Compared to virgin PP, with a crystallinity of 41.2%, the composite with no MA-g-PP displayed slightly lower crystallinity. However, the composites containing 3 and 5 wt.% MA-g-PP exhibited greater crystallinity. Overall, the crystallization rate and the crystallinity of a multi-phase composite depend on the competition between the heterogeneous nucleation provided by the reinforcements and the obstruction they cause during the crystal growth [30,31]. So, for the PPF/MA0/R300 sample, the presence of WCFF did cause an obstruction rather than acting as an effective nucleating agent. However, adding MA-g-PP provided a better dispersion of WCFF within the matrix and an enhanced affinity between WCFF and the PP matrix, and thus, the nucleating effect was more dominant.

As Figure 5d-f shows, the composites processed with 300, 600, and 900 rpm reported similar T_c values of 121.1, 121.7, and 120.7 °C, and crystallinity values of 46.8, 46.7, and 41.6 %, respectively. The increase of screw speed from 300 to 600 rpm did not affect the crystallinity significantly; however, at 900 rpm, the crystallinity decreased slightly. Probably, the highest screw speed broke the WCFF fibers and PP chains, negatively affecting the crystallinity, due to smaller fiber sizes, as also reported in [32].

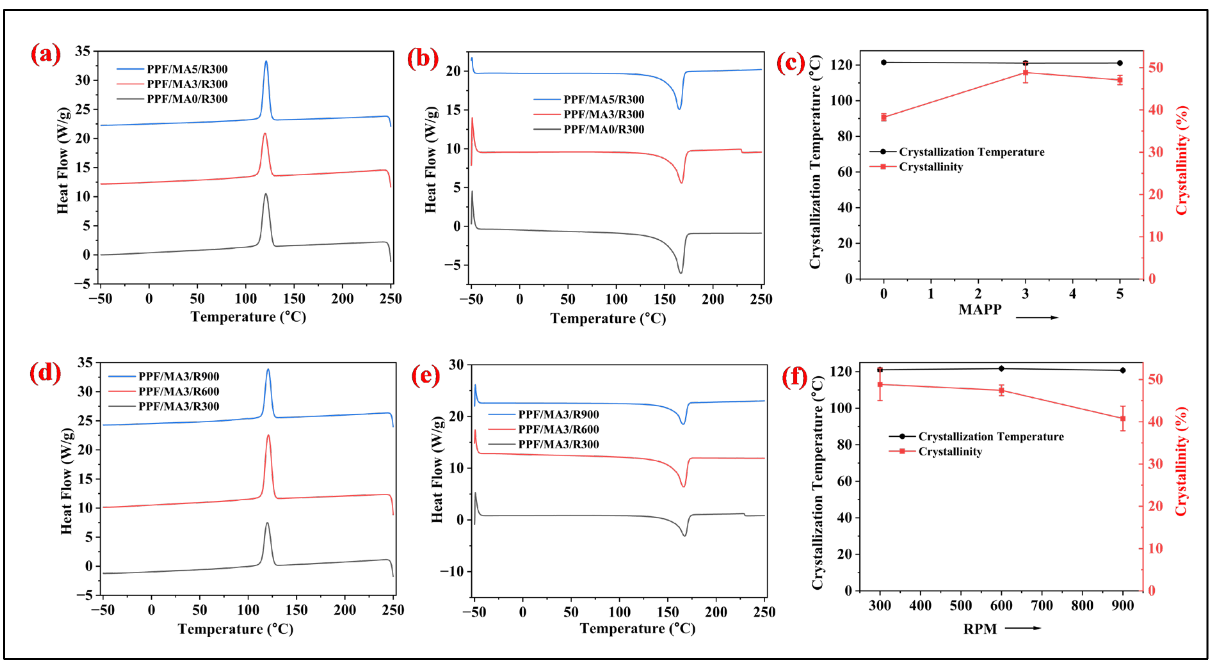

Figure 6 Effect of MA-g-PP on the thermal behavior of PP/20%WCFF composites: (a) cooling thermograms, (b) second heating thermograms, and (c) crystallization temperature and crystallinity. Effect of compounding speed: (d) cooling thermograms, (e) second heating thermograms, and (f) crystallization temperature and crystallinity.

3.5 Density and water absorption

Figure 7a shows the density of the composites at various MA-g-PP content and screw speed, measured on molded samples. Considering all the composites, a grand average of 0.991 g/cm^3 was obtained. The analytical density of the composites was estimated to be 0.996 g/cm^3, using the measured density of virgin PP (0.895 g/cm^3) and the density of cellulose (1.40 g/cm^3) at a loading of 20 wt.%. It was also assumed that MA-g-PP had a similar density to that of PP. The difference between the measured and analytical density values of the composites was only about 0.5%, indicating excellent agreement between the two and evidence of void free samples with the correct dosing of WCFF. Analysis of Variance was also conducted on the effect of MA-g-PP and screw speed on the density with the obtained p-values of $0.237 > 0.050$ and $0.132 > 0.050$, respectively, revealing that there were no statistically significant effect of MA-g-PP and screw speed on the density of the composites at 95% confidence level. These findings are in line with similar reports in [33–35] and [36,37] for MA-g-PP and screw speed, respectively.

Figure 7b also depicts the effect of MA-g-PP content and screw speed on the water absorption of PP/WCFF composites. Overall, the water content was greatly reduced with the MA-g-PP addition but did not change significantly with screw speed. With the addition of 20 wt.% WCFF to PP, a water content of 1.9% was measured in PPF/MA0/R300 sample, which was the highest among all the composites, possibly due to pores or cavities arising from the lack of compatibility between the matrix and the fibers. The water content, however, dropped by about 50% upon introducing 3 and 5 wt.% MA-g-PP. The MA-g-PP improved the interfacial adhesion, thereby reducing the pathways for water to penetrate the material, essentially making the composite more water-resistant. These findings are in line with the previous findings of Yeh and Gupta [38], Korb [39], Zhiltsova et al. [40], and Yatigala [41]. Moreover, water content appeared to have a slightly downward trend with increased screw speed, though not too significant.

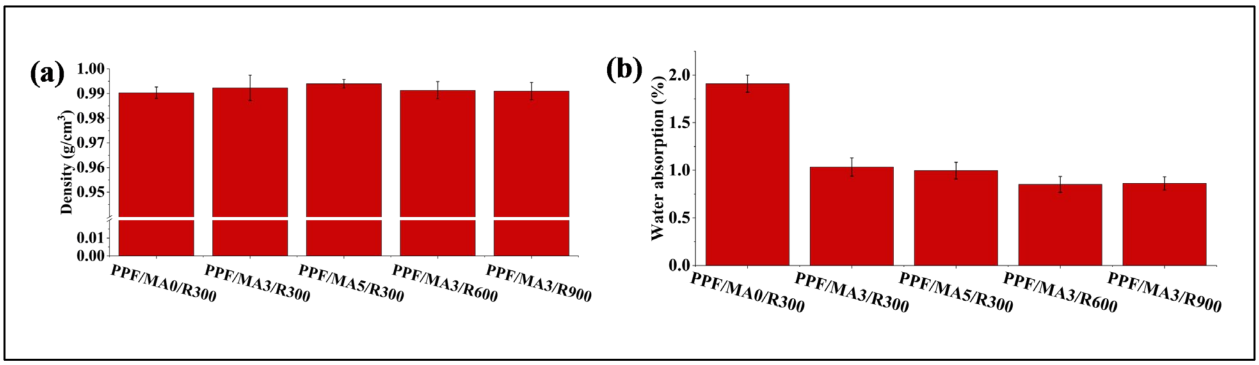

Figure 7 (a) Density and (b) water absorption of the composites at various MA-g-PP contents and screw speeds.

4 Conclusion

This study demonstrated the successful transformation of waste cellulosic fiber and fines (WCFF) from the pulp and paper industry into high-performance polypropylene (PP) composites. Overall, the tensile strength, tensile modulus, crystallinity, and viscosity of the PP/20%WCFF composites were significantly impacted by maleic-anhydride grafted polypropylene (MA-g-PP) content and compounding screw speed. The addition of MA-g-PP at an optimal 3 wt.% content significantly increased the tensile strength and modulus but reduced the elongation at break and viscosity. Compounding screw speed was also found to be critical, with 300 rpm providing optimal fiber dispersion and minimizing fiber breakage and thermal degradation. The PP/20%WCFF with 3 wt.% MA-g-PP processed at 300 rpm resulted in a tensile strength of 41.29 MPa and a modulus of 3,207 MPa, representing improvements of 27.2% and 37.7%, over those of pure unfilled PP. The PP water absorption was significantly increased with the addition of WCFF, about four times; however, the addition of MA-g-PP increased the water resistance by about two times. Moreover, no significant change of water absorption was observed with a change in the screw speed. Overall, the findings highlight that optimized formulations and processes can effectively enhance the performance of PP composites with waste cellulosic materials, contributing to more sustainable composite solutions.

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