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The Influence of MAPP and MAPE Compatibilizers on Physical and Mechanical Properties of 3D Printing Filament Made of Wood Fiber/Recycled Polypropylene

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ABSTRACT

This study aims to develop 3D printing filament composites that support sustainability and waste reduction goals by utilizing wood waste and recycled polypropylene. This study evaluated the effect of Maleic Anhydride Polyethylene (MAPE) and Maleic Anhydride Polypropylene (MAPP) compatibilizers on the mechanical properties of the filament. The study found that r-WoPPc filament with MAPP and MAPE had higher tensile strength compared to r-WoPPc with significant increments of 13% and 74%, respectively, compared to v-WoPPc. The flexural strength of r-WoPPc increased by 18% and 60% after adding optimum loading MAPP and MAPE, respectively. The finding also reveals a significant enhancement in the tensile and flexural strength of the composite, proportional to the increase in MAPP percentage. In contrast, as the MAPE content increases, the tensile strength and flexural strength of the r-WoPPc experience a gradual decrease. Consequently, the addition of MAPP and MAPE improved the interfacial adhesion between wood and polypropylene, as revealed by the surface morphology of the r-WoPPc tensile fractured surface. Moreover, the reduced water absorption in r-WoPPc is attributed to the enhanced interfacial adhesion between wood fibers and the r-PP matrix, associated with improved tensile and flexural strength.

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The highest tensile strength of r-WoPPc with MAPP absorbs 14% water, while the lowest tensile strength absorbs 26%. Likewise, the highest tensile strength of r-WoPPc with MAPE absorbs only 0.8% water, compared to the lowest strength, which absorbs 2% water. This study demonstrated the potential for producing 3D printing filament from recycled polypropylene and wood waste, which benefits sustainability.

Keywords: 3D printing, compatibilizer, filament, wood fiber, recycled polypropylene, MAPP, MAPE, mechanical properties

INTRODUCTION

3D printing has experienced remarkable advancements in recent years, revolutionizing various industries and enabling the production of complex objects with great precision and efficiency. One area of particular interest is the development of 3D printing filament using sustainable and environmentally friendly materials. In this study, an investigation on the influence of compatibilizers, namely Maleic Anhydride Polypropylene (MAPP) and Maleic Anhydride Polyethylene (MAPE), was conducted on the performance and properties of wood fiber/recycled polypropylene filament for 3D printing. In recent years, investigating the effect of maleic anhydride compatibilizer agents on composite properties has gained significant attention in enhancing composite strength. Consequently, this study extends these principles in manufacturing 3D printing filament as they play a crucial role in optimizing the performance of the wood fiber/recycled polypropylene composite filament.

The utilization of wood fiber as reinforcement in polymer composites has gained significant attention due to its desirable properties, such as low cost, abundance, and excellent mechanical performance. On the other hand, recycled polypropylene addresses the crucial concern of plastic waste management while offering favorable thermoplastic characteristics. However, the inherent incompatibility between wood fiber and polypropylene matric poses challenges in achieving optimal interfacial adhesion and mechanical strength in the composites. Typically, wood fiber exhibits a polar nature, while polymer matrices tend to be non-polar (Zhang et al., 2019). Nonetheless, the introduction of the maleic anhydride coupling enhancing the compatibility between the wood fiber and the polymer matrix, which improved adhesion between these two constituents, resulted in better mechanical properties (Bütün et al., 2019; Khalid et al., 2021). The stronger adhesion primarily resulted from forming hydrogen bonds and covalent linkages between wood fibers and maleic anhydride. Subsequently, the hydroxyl group of the wood fibers establishes a stronger bond with the oxygen atoms in the polymer matrices, further enhancing the adhesion (Billah et al., 2022).

Compatibilizers, such as MAPP and MAPE, have become promising additives that enhance the compatibility and interfacial bonding between materials in polymer composites. Reactive functional groups in these compatibilizers facilitate molecule interaction between wood fiber and polypropylene, which promotes adhesion. Compatibilizers effectively bridge the interfacial gap between reinforcing wood fiber and polypropylene matrix strong adhesion, resulting in improved mechanical properties of the composites (Aida et al., 2021). The addition of maleic anhydride as a compatibilizer in PP-based composite during the FDM filament extrusion process also improved the friction and wear rate of the PP composite during the extrusion process, improving the mechanical properties of extruded filament (Kristiawan et al., 2021).

This paper aims to investigate the influence of MAPP and MAPE compatibilizers on the physical and mechanical properties of the wood fiber/recycled polypropylene composite (r-WoPPc) filament for 3D printing. The wood dust was pretreated using silane to ensure better enhancement of the wood fiber and polypropylene (PP) matrix in this study. Silanes are efficient coupling experts commonly utilized in fiber-reinforced composites; by forming compound linkages between the silane and the natural fiber surface, the surface morphology of natural fibers was altered (Khalid et al., 2021). The increase in strength is due to silanol (Si-OH) groups forming, creating strong bonds with the –OH groups of the fibers. For the remaining Si-OH groups, condensation occurs with adjacent Si-OH groups (Atiqah et al., 2018). A study by Petchwattana et al. (2019), who developed 3D printing filament using PLA/teak wood flour composite, encountered that composite filament with wood silane treatment had higher tensile strength compared to those without silane treatment. The impact of two different compatibilizers on the printed parts' tensile and flexural strength was assessed. Furthermore, the study also evaluated the effect of compatibilizer on water absorption of the filament and surface characteristics of the filament. Hence, this research could provide valuable insight into the potential of developing sustainable filament materials for 3D printing.

Literature Review

Maleic coupling agents are commonly employed to enhance the properties of polymer composites that incorporate filler or fiber reinforcement (Khalid et al., 2021). The incompatibility between these two constituents can be resolved by reacting anhydride groups in malaeted polymer additive and the hydroxyl group of natural fibers. This interaction enhances the tensile properties of the composites. Generally, natural fibers, including wood, are polar, and polymer matrices are non-polar (Zhang et al., 2019). Hence, Maleic Anhydride Polypropylene (MAPP) and Maleic Anhydride Polyethylene (MAPE) are commonly used as polymer blend compatibilizers to improve mechanical properties and compatibility. When used as compatibilizers in 3D printing, fused deposition modeling (FDM) filaments made of wood fiber/recycled polypropylene can influence the properties of the resulting material. The MAPP and MAPE help in improving the compatibility between the polar surface of wood fiber and the non-polar surface of the PP matrix (Bütün et al., 2019; Khalid et al., 2021).

To begin with, the addition of MAPP and MAPE can improve the adhesion of wood fiber to recycled polypropylene. It can result in a more uniform distribution of the wood fiber within the polymer matrix, improving the mechanical properties of the resulting material. Improved adhesion reduces the likelihood of delamination during printing, improving the printed parts' quality (Razak et al., 2018). MAPP was used as a compatibilizer to competently enhance fiber-matrix bonding because of the formation of hydrogen bonds and covalent linkages between the hydroxyl groups of the fiber and maleic anhydride. The hydroxyl group of wood fiber, a chemical functional group of lignin, creates a strong hydrogen bond with the oxygen atom of MAPP (Billah et al., 2022).

Composite reinforced with wood is generally developed using raw wood fiber without any treatment or modification. Wood is alkali-treated to remove impurities, enhance properties, or improve compatibility with the matrix material. Wood is impregnated with a compatibilizer that helps to improve its compatibility with the matrix material. Also, silane is a coupling agent that enhances interfacial adhesion between fiber and matrix. The purpose of using these compatibilizers is to enhance the overall performance of the composites (Baykus et al., 2016). Keener et al. (2004) mentioned that adding maleated compatibilizing agents with a balance of maleic anhydride and molecular mass helps provide composites with the best performance. This is due to its function of enhancing the effectiveness of the oxidation of the polymer matrices with the fiber (Hao et al., 2021). Their work found that adding 1%–5 % MAPE or MAPP to the WPC improved mechanical properties by 30%–100 %.

Maleated coupling agents are commonly preferred in the literature for various polymer and fiber systems. Although maleated coupling agents are most utilized directly during the mixing process, they can also be grafted on hydroxyl groups of fiber or pre-impregnated with fiber prior to manufacturing. The physical and mechanical features of the wood particles, as well as the chemical interaction between wood particles and polymers, affect the quality of WPCs. Couplers like maleated anhydride grafted polyethylene and maleated anhydride grafted polypropylene are commonly used to improve the compatibility between hydrophilic cellulosic materials and a hydrophobic polymer (Mu et al., 2018). Moreover, high compatibility aids particle dispersion and adequate wetting in matrices, resulting in strong contact adhesion. Because ester linkages promote fiber-matrix contact, stress can be transferred more easily from the matrix to the reinforcing particles, improving mechanical characteristics. Furthermore, adequate contact adhesion lowers WPC water absorption (Dhanalakshmi et al., 2017).

MATERIALS AND METHODS

The wood dust used in this study was sourced from a local furniture workshop. The wood dust was treated using silane ((3-aminopropyl) triethoxysilane with CAS 919-30-2) from

Sigma-Aldrich. The recycled PP, a byproduct of plastic products, was supplied by a plastic factory in Ayer Keroh, Melaka.

Preparation of r-WoPPc

The wood dust was sieved to obtain 125 µm size, thoroughly washed with distilled water to eliminate impurities, and then dried in a hot air oven at 60°C for 24 hours. The wood dust was soaked in 2 wt% silane solution for 3 hours and dried at 80°C for 72 hours (Atiqah et al., 2018). The sample of treated wood dust was ready to mix with polypropylene after three days in the oven. The loadings of MAPP and MAPE used were 1%, 3%, and 5% respectively. Meanwhile, wood dust loading was fixed at 3%.

Silane-treated wood dust was mixed with recycled PP pellets, pressed using a hot press, and later crushed into pellets prior to making the filament. r-WoPPc pellet was then fed into a single screw extruder to produce filament. The filament of the composite material was extruded using a single screw extruder equipped with a die nozzle of 1.75 mm. The screw speed was set into range (18–20 rpm). To produce a 3D printed product, the dumbbell for tensile and flexural models, according to ASTM D638 and ASTM D790, was made using SolidWorks Software. The 3D modeling files were converted into STL files, which were later printed using the Ender 3D printing machine, as shown in Figure 1.

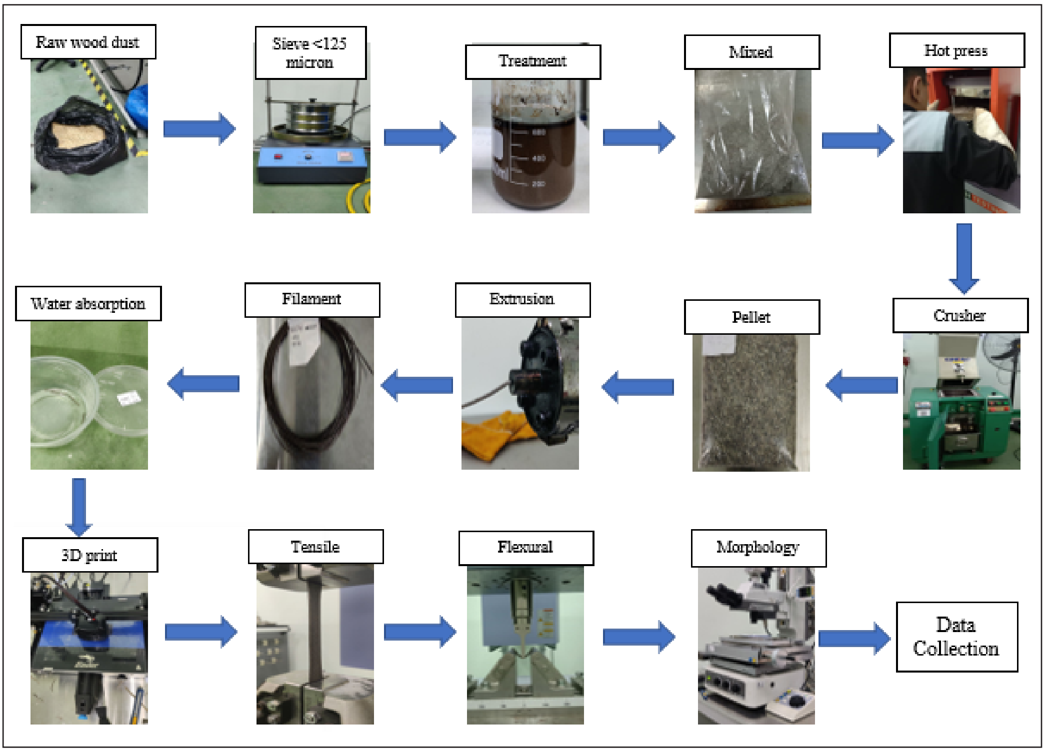


Figure 1. Production of 3D printing filament of r-WoPPc

Tensile and Flexural Tests of r-WoPPc

Tensile and flexural tests were conducted in accordance with ASTM D638 and ASTM D790 standards using Shimadzu AGS-X Universal Testing Machine. The following conditions were used: 5 mm/s displacement with a 30mm/min crosshead speed.

Surface Morphology r-WoPPc Tensile Fracture Surface

Surface morphological analyses of the tensile fractured surface were conducted using a Nikon MM-800N high-precision microscope.

Water Absorption of r-WoPPc

The water absorption of r-WoPPc was evaluated according to ASTM D570-98 to assess the hydrophilicity of the composite. The r-WoPPc filament was cut into 20 mm lengths and soaked in distilled water for 30 days. The weight of samples was recorded before and after immersion using the analytical balance model JA203P with ± 0.002 g accuracy. The water absorption of the composites was determined using Equation 1.

$$Water\ Absorption\ (\%) = \frac{W_1 - W_0}{W_0} \times 100\% \quad (1)$$

where w_o (g) is the weight of composites after drying and w_1 (g) is the weight after water immersion.

RESULTS AND DISCUSSION

The mechanical properties of 3D printed parts are crucial for their function performance in various applications. This study investigated the influence of MAPP and MAPE compatibilizers on the tensile and flexural strength of 3D printed parts made of wood fiber/recycled polypropylene filaments. Polypropylene matric with low melt viscosity can penetrate the wood fiber cellulose, which decreases the number of voids. Fewer voids resulted in higher density composite with improved mechanical properties (Yuan et al., 2008). Adding maleic anhydride polypropylene or polyethylene as a compatibilizer significantly enhanced the interfacial adhesion between the wood fiber and the polypropylene matrix. As a result, the composite strength improved considerably (Aida et al., 2021).

Effect of MAPE Compatibilizer Concentrations on Mechanical Properties

Figure 2 shows the variation of tensile strength with different compatibilizer concentrations. It can be observed that the addition of MAPE improved the tensile strength of the 3D-printed part produced from the r-WoPPc filament. The tensile strength of the r-WoPPc composite with MAPE compatibilizer was 74% higher than without compatibilizer. The composite's tensile strength was improved due to the enhanced bonding between the wood fiber and the polypropylene matrix facilitated by the compatibilizer. Compatibilizer helped create stronger connections at the interface between the wood fiber and the polypropylene, leading to an overall enhancement in the mechanical properties of the composite. Introducing the compatibilizer into the mixture initiates the reaction of the anhydride group with the alcohol hydroxyl group within the wood fiber, thereby decreasing the fiber's polarity and hydrophilicity. This reduction in polarity strengthens the compatibility between the wood fiber and polypropylene.

Additionally, this process improves compatibility between the wood fiber and polymer surfaces. However, a higher compatibilizer concentration beyond the optimum point decreased tensile strength, indicating a possible plasticization effect. It can be observed that when loading MAPE was further increased to 3 wt% and 5 wt%, the tensile strength of the composite started to decrease, indicating the optimum MAPE value for the r-WoPPc 3D filament was 1 wt%. This finding was comparable with a study by Zhou et al. (2022), as adding MAPE into the wood fiber composite improved the tensile and flexural strengths because MAPE enhanced the interfacial adhesion between wood fiber and polymer matrix. This resulted in efficient stress transfer from the polymer to the wood fiber (Mu et al., 2018; Ou et al., 2014).

This study assessed the flexural strength of the 3D printed parts fabricated using r-WoPPc filament to measure their resistance to bending and deformation. Figure 3 shows the relationship between the flexural strength of the WoPPC 3D-printed part and compatibilizer concentration. Like the tensile strength, the flexural strength improved by adding MAPE. MAPE promoted better stress transfer between the wood fiber and polypropylene, resulting in increased flexural strength of approximately 60% than without a compatibilizer. However, the addition of excessive amounts of compatibilizer at concentrations of 3wt% and 5wt% showed decrement in tensile strength, indicating 1% as the optimum value of MAPE for the r-WoPPc filament.

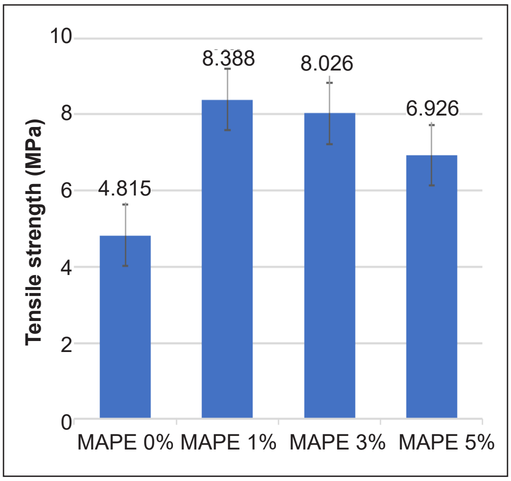


Figure 2. Tensile strength of r-WoPPc with MAPE

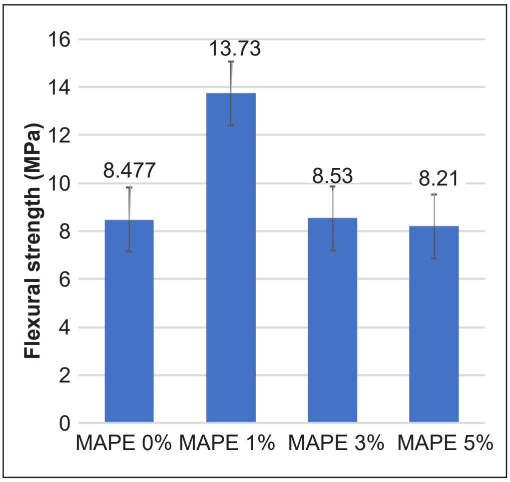


Figure 3 Flexural strength of r-WoPPc with MAPE

Effect of MAPP Compatibilizer Concentrations on Mechanical Properties

Figure 4 illustrates the effect of varying percentages of MAPP on the tensile and flexural strength of the r-WoPPc composite part. Notably, the introduction of MAPP led to an enhancement in the tensile strength of the composite by 13% compared to v-WoPPc. Furthermore, the tensile strength of the composite increased proportionally with a higher percentage of MAPP compatibilizer. The result indicates a notable enhancement in the tensile strength of the r-WoPPc as the MAPE content increased to 3 wt% with an increase of tensile strength from 13% to 20%. Furthermore, the highest amount of 5wt% MAPP resulted in a remarkable 40% increase in tensile strength. However, the rWoPPc flexural strength decreased by adding MAPE of 1 wt%. However, it improved proportionally with the addition of 3 wt% and 5 wt% of MAPP, with 5 wt% MAPP resulting in the highest flexural strength with an increment of 17.8% compared to v-WoPPc (Figure 5). It is concurrent with a study by Silva et al. (2021) and Petchwattana et al. (2019).

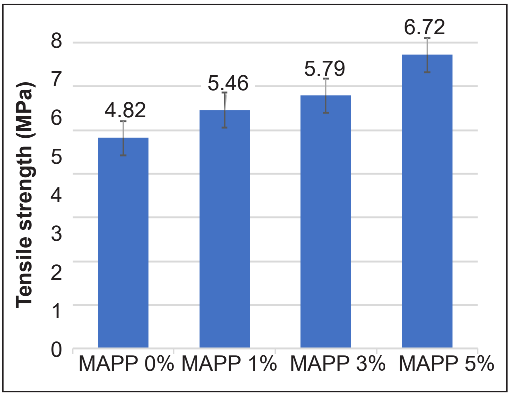


Figure 4. Tensile strength of r-WoPPc with MAPP

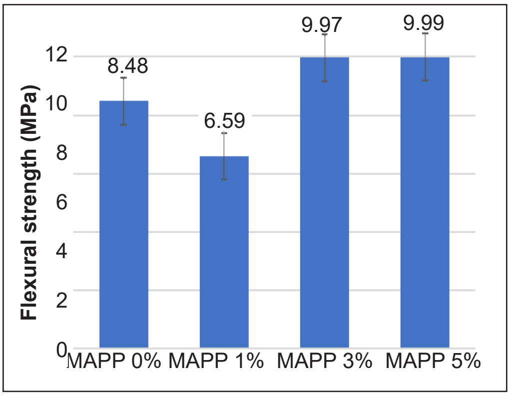


Figure 5. Flexural strength of r-WoPPc with MAPP

Effects of MAPE Compatibilizer on r-WoPPc Tensile Fracture Surface and Filament Surface Morphology

Figure 6 shows the SEM micrograph of the r-WoPPc tensile fracture surface with various MAPE loadings. In the absence of MAPE compatibilizer, there was inadequate adhesion between the wood fibers and the r-PP matrix, resulting in the wood fiber easily fractured and pulled out from the matrix (Figure 6a). Meanwhile, Figure 6(b) demonstrates strong adhesion between the wood fiber and r-PP matrix when MAPE was incorporated in r-WoPPc. The evidence of the wood fiber impregnated well in the r-PP matrix was enhanced interfacial bond, resulting in higher tensile and flexural strengths (Hao et al., 2021). Contrary to this, incorporating higher quantities of MAPE did not improve adhesion between the wood fiber and r-PP matrix, as evidenced by the SEM micrographs. Figures 6(c) and (d) show poor interfacial adhesion between the wood fiber and PP matrix in the r-WoPPc was incorporated with 3 wt% and 5 wt% MAPE.

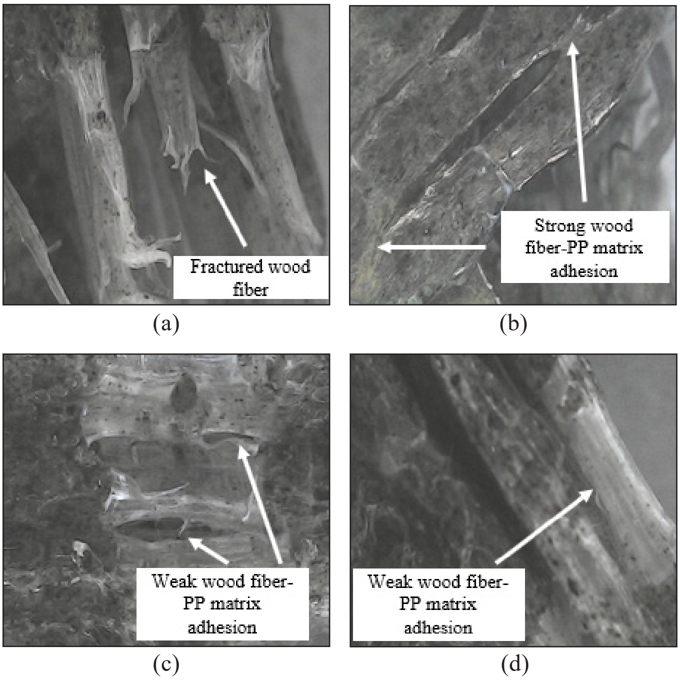


Figure 6. High precision micrograph with 10× magnification of r-WoPPc tensile fracture surface: (a) without MAPE; (b) 1 wt% MAPE; (c) 3 wt% MAPE; and (d) 5 wt% MAPE

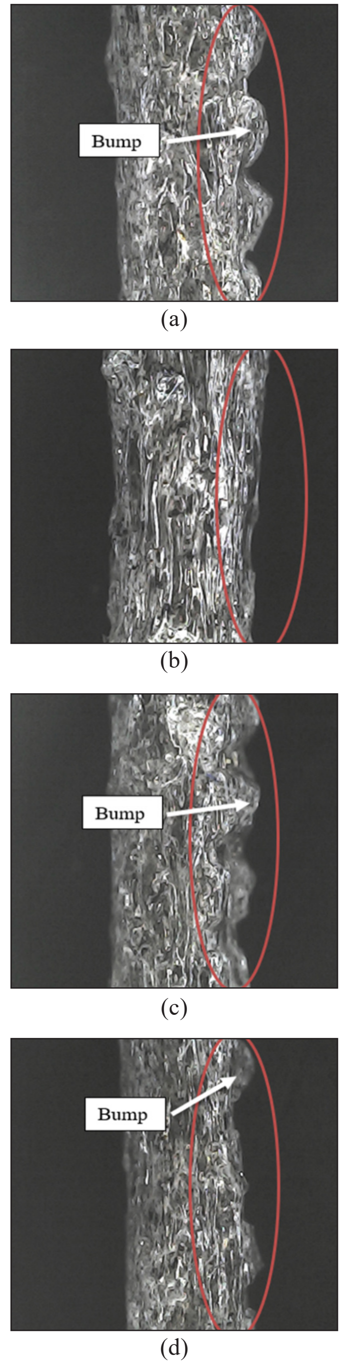


Figure 7. r-WoPPc filament surface morphology: (a) without MAPE; (b) 1 wt% MAPE; (c) 3 wt% MAPE; and (d) 5 wt% MAPE

Figure 7 shows the filament surface morphology with various MAPE loadings taken using a high-precision microscope. For filament without MAPE in Figure 7(a), the filament's surface was uneven, possibly lacking bonding between wood and r-PP matrix. Meanwhile, r-WoPPc with 1 wt% MAPE produced a smooth and even filament, as shown in Figure 7(b). However, adding more than 1 wt% loading of MAPE seemed to tamper the optimum interfacial adhesion between wood and r-PP, as the filament surface appeared uneven and not smooth, as shown in Figures 7(c) and (d) for r-WoPPc with 3 wt% and 5 wt% MAPE, respectively.

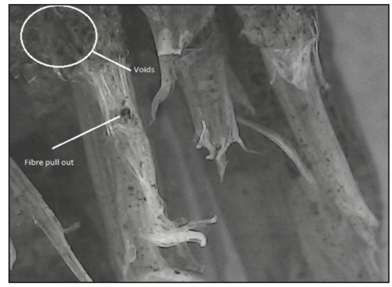
Effects of MAPP Compatibilizer on r-WoPPc Tensile Fracture Surface Morphology

Similarly, incorporating MAPP into the r-WoPPc improved the interfacial adhesion between the wood fiber and the r-PP matrix, much like the effect observed with MAPE. Figure 8 shows the r-WoPPc filament surface morphology without

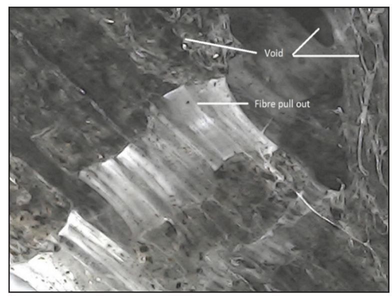
MAPP (0 wt%), with 1 wt%, 3 wt%, and 5 wt% of MAPP loadings, respectively. Figure 8(a) demonstrates that the absence of MAPP in the r-WoPPc composite affected the integrity of the composite; having more fiber pull-out led to more holes (Çavuş, 2020). A noticeable wood fiber rupture and fiber pull-out from the matrix were observed. The surface morphology of r-WoPPc with 1% MAPP showed slightly better adhesion compared to composite without MAPP, showing that compatibilizer helped enhance the wood and PP matrix (Amir et al., 2017). Meanwhile, it can be observed that the r-WoPPc containing 3 wt% MAPP loading, as depicted in Figure 8(c), showed better interfacial adhesion between wood and PP.

Moreover, with 5 wt% MAPP loading, as shown in Figure 8(d), significantly improved interfacial adhesion between the wood and r-PP matrix, which demonstrated the highest tensile and flexural strengths of the r-WoPPc were found. Additionally, 5 wt% MAPP significantly enhanced the composites' interfacial adhesion between wood and r-PP matrix. This improvement reduced void formation, effectively minimizing stress concentration points that could potentially compromise the strength of the composites (Sosiati et al., 2018). Adding maleic anhydride as a compatibilizer in PP-based composite also improved PP's friction and wear rate during the FDM extrusion process, which produced improved mechanical properties of the extruded part (Kristiawan et al., 2021).

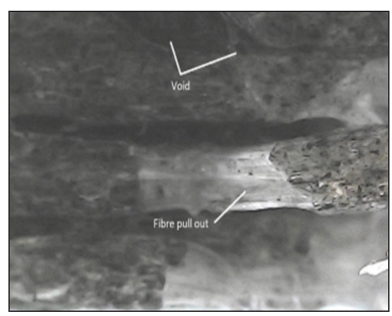
Effects of MAPE Compatibilizer on Water Absorption of r-WoPPc. The amount of water uptake or absorption in the composite indicates the composite structural integrity. Higher water absorption is associated with more voids in the composites, leading to a detrimental effect on the composite mechanical properties. Thus, the quantification of water uptake in the r-WoPPc provided significant information regarding the presence of voids and their impact on the tensile and flexural strengths. A lower number of r-WoPPc water



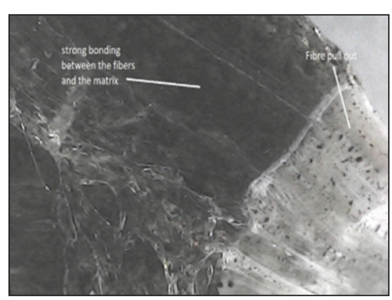
(a)



(b)



(c)



(d)

Figure 8. High precision micrograph with 10 × magnification of r-WoPPc tensile fracture surface: (a) without MAPP; (b) 1 wt% MAPP; (c) 3 wt% MAPP; and (d) 5 wt% MAPP

absorption signified that the r-WoPPc had a more compact structure and fewer voids, typically associated with improved tensile and flexural strengths. In contrast, voids were present when r-WoPPc absorbed a higher amount of water. These voids caused detrimental effects on the tensile and flexural strengths due to additional stress concentration points.

The water absorption of r-WoPPc with different MAPE loadings provides significant proof of the influence of compatibilizer on composites' structure. Figure 9 presents the water absorption result of r-WoPPc. It is apparent from the data that the incorporation of 1 wt% MAPE in r-WoPPc led to significantly reduced water absorption. The r-WoPPc with 1% MAPE absorbs a smaller amount of water, which was 0.8%, compared to a higher absorption rate for 3 wt% MAPE addition and 5 wt% MAPE addition, which are 2% and 1.1% water absorbed, respectively. The lower water absorption value of r-WoPPc was attributed to the improved interfacial adhesion between the wood fiber and PP matrix, which signified the composite's tensile and flexural strength. This result was similar to the finding of (Zhou et al., 2022). These water absorption results aligned with the tensile and flexural strengths of the r-WoPPc composites with 1 wt% MAPE, obtaining the highest strength among all tested composites. Meanwhile, water absorption of composites containing 3 wt% MAPE was higher than 1 wt% and 5 wt% MAPE loadings, which evidenced that the composite had more voids compared to the composite with 1 wt% MAPE. The reduction in water absorption of the r-WoPPc was confirmed by the morphology results that showed strong adhesion between the wood and r-PP matrix.

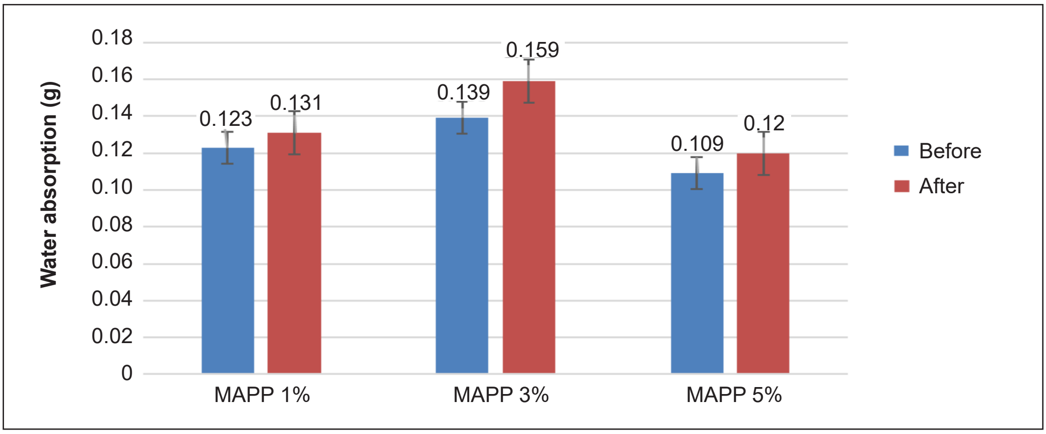


Figure 9. r-WoPPc water absorption values with varying MAPE loadings

Effects of MAPP Compatibilizer on Water Absorption of r-WoPPc

The study of water absorption in r-WoPPc with varying MAPP loadings provides substantial evidence of the influence of the compatibilizer in improving interfacial adhesion between wood and PP, as shown in Figure 10. Composite with 5 wt% MAPP absorbed the least water compared to 1 wt% and 3 wt% MAPP, respectively. This finding validated the smaller number of voids in the composites, as observed in filament morphology resulting from improved interfacial adhesion of wood and PP, subsequently improved composite strength. The finding was similar to the study conducted by Petchwattana et al. (2019), who showed that using a compatibilizer significantly reduced water absorption compared to without a compatibilizer (Billah et al., 2022).

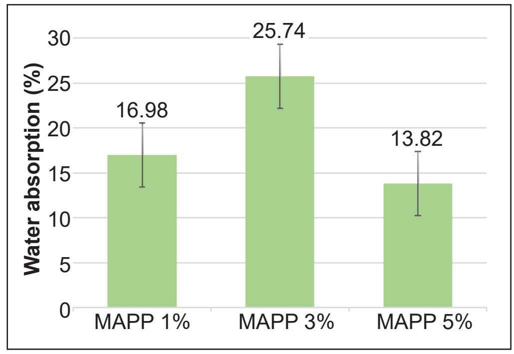


Figure 10. r-WoPPc water absorption values with varying MAPP loadings

CONCLUSION

In conclusion, adding MAPP and MAPE compatibilizers improved the mechanical properties of the wood fiber/recycled polypropylene (r-WoPPC) 3D printing filaments. The addition of MAPP improved the tensile and flexural strength of the composite by 13% and 18%, respectively, and the addition of MAPE enhanced the composite tensile and flexural strength by 74% and 60%, respectively. This study identified that the optimal MAPP content for achieving the highest mechanical performance of the r-WoPPc was 5 wt%, whereas for MAPE, it was 1 wt%. A significant improvement in r-WoPPc mechanical performance was observed as the percentage of MAPP in the composite increased. The tensile and flexural strength increased proportionally with the percentage of MAPP amount.

In contrast, as the MAPE percentage increases, the mechanical performance of the composite decreases gradually. This observation is likely attributed to the nature of the coupling agent-based and their compatibility with the polypropylene matrix. As PP- a based compound- MAPP provides more effective intermolecular or intramolecular coupling between the r-PP molecules and wood fibers. Therefore, increasing the MAPP percentage strengthens the interfacial adhesion between the wood fibers and the r-PP matrix. This finding highlights the importance of selecting a suitable coupling agent for achieving desired composite performance. Moreover, the increasing proportion of maleic anhydride coupling agents is shown to enhance the structural integrity of the composite, provided that the basis of the coupling agent is compatible with the composite matrix. The enhanced mechanical properties of r-WoPPc 3D printing filament were ideal for prototyping parts and products to produce architectural and structural models, furniture, and consumer goods.

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