

Proceeding Paper

Transforming Agricultural Waste into Sustainable Composite Materials: Mechanical Properties of Tamarindus Fruit Fiber (TFF)-Reinforced Polylactic Acid Composites ⁺

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Abstract: Natural fiber-based polymer composite has great potential and is in high demand due to its high specific strength, low carcinogenic nature and economic market value. Tamarindus Fruit Fiber (TFF) has low density, high tensile strength and is easily available. It is one of the industrial waste materials. Polylactic acid (PLA) possesses an entangled coherence with fiber, since it is more compatible. A sample was prepared by integrating TFF as fiber and PLA as the binding agent. The fiber variations in all samples were 10 to 50 wt.% step by 10 wt.%. A pure PLA sample was also fabricated for the purpose of comparison. Mechanical properties such as tensile strength, flexural strength, impact and hardness have been evaluated. It was revealed that TFF reinforcement increased the mechanical properties of the samples. The highest mechanical properties were observed in Sample S5, which had 40 wt.% TFF and 60 wt.% PLA. Fracture failure was found using fractographic analysis. In conclusion, this study demonstrates the potential of utilizing TFF as an agricultural waste product for enhancing the mechanical properties of biodegradable polymer composites. These sustainable compositions of materials have been used for many applications in various industries, including packaging, automotive, and construction, while also providing an environmentally friendly solution for agricultural waste products.

Keywords: polylactic acid; tamarindus fruit fiber; mechanical characterization; scanning electron microscope; agricultural waste; structural applications

1. Introduction

Agricultural residues account for approximately 20% of global agricultural production, which translates to approximately 2.2 billion tons of waste per year [1]. In recent years, there has been an increasing interest in utilizing agricultural waste as a source of natural fibers for reinforcing polymer resins because of their availability, low cost and potential environmental benefits [2]. Several agricultural waste products, including rice husks, sugarcane bagasse, wheat straw, and coconut fibers, have been investigated for the purpose of replacing glass fiber in various industries, since these fibers are available naturally and are stronger when combined with resin [3]. The mechanical properties of these composite materials can vary widely depending on the type of fiber, processing method and polymer matrix used [4]. One of the challenges in utilizing agricultural waste for reinforcement of polymer resins is the variability in the fiber properties, which can affect the mechanical properties of the resulting composites. However, with the careful selection of fiber sources and processing methods, agricultural waste fibers have the potential to enhance the mechanical properties of polymer composites and provide a sustainable solution for waste management [5–10].



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In recent years, significant research has been conducted on the utilization of agricultural waste for reinforcement of polymer composites. Rice husk, sugarcane bagasse, wheat straw and coconut fibers are among the agricultural waste products most commonly used for this purpose [11–15]. Many researchers have the thought of preparing natural fiber-reinforced polymer composite with an affordable price and good strength, considering the proper manufacturing method for the specific fiber and polymer, since it varies based on types of filler and resin [16–20]. High-density polyethylene particulate filled with rice husk provides significant tensile and flexural strength while increasing the filler content, but the impact strength is reduced [21–24]. Similarly, in a study, it was proved that the mechanical properties of the sugarcane bagasse fiber-reinforced polystyrene (PS) composites were increased by increasing the fiber content. A similar finding was found in many other papers [25–30].

The problem statement for this study was that agricultural waste materials are often underutilized and can contribute to environmental pollution if not properly managed. The novel statement is that using tamarindus fruit fiber (TFF) as a reinforcement material in polymer composites has not been extensively studied, despite its abundance and potential as a sustainable material. The aim of this work was to improve the performance of PLA resin by reinforcing the fiber TFF. The mechanical properties of this prepared composite (TFF-reinforced PLA composites) were identified with the aim of developing a sustainable and biodegradable material with improved mechanical performance. Figure 1 shows the description of all the levels throughout this study. Specifically, this study aimed to determine the optimal TFF loading percentage and processing conditions to achieve maximum properties of the resulting composite materials.



Figure 1. Process flow chart.

2. Materials and Methods

2.1. Materials

Polylactic acid (PLA) resin with a grade of 4043D was obtained from Leo Enterprise, Nagercoil. The tamarindus fruit fiber (TFF) was sourced from local agricultural waste. The melting temperature of PLA is 150–160 °C and the density is 1.24 g/cm³. It is a biodegradable polymer with good mechanical properties that is commonly used in sustainable packaging applications. The fiber has a diameter of 25 μ m. The length varies from 500 to 1000 μ m. The peeled fiber was primarily composed of cellulose (approximately 67%) and hemicellulose (approximately 25%), with smaller amounts of lignin and pectin. This fiber has a high aspect ratio and good mechanical strength, making it a promising reinforcement material for polymer composites [31–33].

2.2. Methods

A compression molding technique was employed to fabricate TFF-reinforced PLA composite samples. Initially, PLA resin and TFF fibers were mixed in a rotary mixer at a constant speed of 60 rpm for 10 min to ensure proper fiber dispersion within the resin. Subsequently, the mixture was compressed at 170 °C and 10 MPa using a hydraulic press for 5 min, resulting in 2 mm thick sheets, which were then cut into dumbbell-shaped samples labeled as S1 to S6, representing varying fiber percentages from 0 to 50 wt.%. The ASTM D638 standard was followed for tensile testing, which was conducted using an Instron—UTM machine with a cross speed of 2 mm/min [4]. Three-point bending tests for flexural strength and modulus were performed on 40 mm gauge length samples at a crosshead speed of 2 mm/min, adhering to the ASTM D790 standard [11]. Additionally, shock load energy absorption behavior was assessed in accordance with ASTM D256 [12]. Surface hardness was determined using a Shore D durometer following ASTM D2240 guidelines [28]. These tests were carried out to evaluate the composite specimens across a range of TFF loading percentages, ultimately leading to the determination of optimal processing conditions based on preliminary experiments.

3. Results and Discussion

3.1. Performance on Tensile Strength and Modulus

The tensile properties of the prepared samples have been tested with the aid of Instron—UTM. The computed values were analyzed and compared to find the optimum level, which is shown in Figure 2. The addition of Tamarindus Fruit Fiber (TFF) significantly improves the performance of the materials. The unfilled pure resin (S1) has tensile strength of 26.19 MPa and the TFF (40 wt.%) reinforced PLA composite (S5) has tensile strength of 54.45 MPa, which is more than 100% of the pure sample. Similarly, the tensile modulus increased from 1.24 GPa for the pure PLA to 1.79 GPa for the composite containing 40 wt.% TFF. The enhancement in the mechanical properties can be attributed to the reinforcing effect of TFF, which is due to its high aspect ratio and stiffness.



Figure 2. (a). Tensile strength of PLA/TFF composite. (b) Tensile modulus of PLA/TFF composite.

The sample with more than 40 wt.% of TFF (S6) showed a drop in tensile strength and tensile modulus because of inadequate resin in the sample, which means the bonding between the fiber and the resin reduced simultaneously. The tensile strength decreased to

46.23 MPa and the tensile modulus decreased to 1.53 GPa. The voids also increased when the TFF increased in the sample due to the hydrophobic nature of the fiber. The formation of the oxide layer begins in this stage.

3.2. Performance on Flexural Strength and Modulus

The bending strength and modulus of the samples are analyzed in Figure 3. The value was increased in both of the graphs by adding TFF up to a certain level; after that, the phenomenon changed. This can be attributed to the reinforcement effect of the TFF fibers in the matrix, which helped with the distribution of the load across the composite structure, resulting in a higher resistance to bending. Samples S4 and S5 exhibited the highest flexural strength and modulus values among all samples because of the optimal concentration of TFF fibers in the composite.



Figure 3. (a). Flexural strength of PLA/TFF composite. (b) Flexural modulus of PLA/TFF composite.

The prepared samples do not have significant value for flexural strength because of resin. Resin does not have a good bending nature. Nevertheless, the reinforcement of TFF increased the flexural strength to 54.45 MPa and the tensile modulus to 2.65 GPa in sample 5, including the fiber percent of 40 wt.%. The maximum tensile strength has a more than 100% hike compared to that of the pure sample and the pure sample exhibited a 34% drop in the tensile modulus compared to maximum value.

3.3. Impact Strength

The impact strength results for the TFF-PLA composite samples are shown in Figure 4. When the amount of fiber increased, the impact strength also increased. The trend increased with the addition of up to 40 wt.% TFF and then showed a slight decrease at 50 wt.% TFF content. The impact strength of S1 (100 wt.% PLA) was found to be the lowest among all the composite samples. The increase in the impact strength of the composites can be attributed to the increased toughness of the TFF particles, owing to their fibrous nature and good interfacial bonding with the PLA matrix. The interfacial bonding between the TFF particles and PLA matrix leads to the effective transfer of stress from the matrix to the particles, thus enhancing the energy-absorption capacity of the composites. However, at higher TFF content (50 wt.%), the decrease in impact strength could be due to the agglomeration of TFF particles, which leads to a non-uniform stress distribution and stress concentration points. Overall, the impact strength of the TFF-PLA composite samples was significantly improved



compared to that of pure PLA. Once the potential of TFF increased, the sustainability of the material increased for high-performance composites.

Figure 4. Impact strength of PLA/TFF composite.

3.4. Hardness

The hardness of the PLA/TFF composites increased with the increase in TFF content (Figure 5). This trend was attributed to the incorporation of TFF, which could have acted as a filler and increased the density of the composites. The sample with the highest TFF content (S5) had the highest hardness value of 69 Sd, which was significantly higher than that of the pure PLA sample (S1), which had a hardness value of 54 Sd. The increase in the hardness values of the composites could also be attributed to the strengthening effect of the TFF, which contributed to a higher resistance to indentation. The results suggest that the addition of TFF could enhance the hardness of PLA/TFF composites, which could be beneficial in applications that require high wear-resistance and durability. The fiber covers the gap with polymer which prevents penetration and removal of particles from the surface area [34–36].



Figure 5. Shore D hardness of PLA/TFF composite.

3.5. Morphological Study

A scanning electron microscope was used to study the reason for the failure of fractured surfaces [37,38]. It was found that a significant result was obtained in sample Id S5. This means it has a TFF fiber percentage of 40 wt.% and a PLA percentage of 60 wt.%. The fractography of sample Id S5 after the tensile test experiment is shown in Figure 6. The

main fracture mechanics were poor fiber–matrix bonding (Figure 6a) and matric fracture (Figure 6b). However, the reason for the subsequent higher properties is matrix penetration of the fiber cell wall, as shown in Figure 6c. The images substantiate the significant failure of the broken surface, which is mainly due to poor bonding above 40 wt.% fiber loading. That is why the result reaches its maximum at 40 wt.% fiber loading.



Figure 6. SEM analysis of PLA (60%)/TFF (40%) composite (Sample Id.:5). (**a**) Delamination and fiber pull-out zone, (**b**) matrix fracture zone, (**c**) matrix penetration inside the cell wall.

The fiber dispersion is quite good throughout the sample. The matrix penetration is identified inside the cellulose wall. It increases the bonding between the fiber and matrix. It is the main cause of the wetting behavior of the resin. And hence, the matrix traveled to the sandwich portion of the cellulose core. Delamination was also observed via SEM. This emphasizes the chaotic stage of the poor bonding due to the presence of impurities and wax around the outer cellulose wall.

The fiber fracture can also be seen in the image, which shows the utilization of full fiber strength against loads involving proper bonding strength. A random void is also present in the sample, which is a sign of starting failure. Delamination is mainly seen in the reinforced fiber region. A matrix fracture around the fiber is also observed, and is another sign of adhesiveness in the area.

4. Conclusions

Our research focused on developing six different samples to find the optimum level for various properties. The tensile strength and modulus of sample S1 is 26.16 MPa and 1.24 GPa in the pure PLA sample. But the maximum tensile strength (54.45 MPa) and modulus (1.79 GPa) were obtained at 40 wt.% of TFF, and were 108% and 44% higher than those of the pure base sample, respectively. Furthermore, the maximum flexural strength (65.36 MPa) and modulus (2.65 GPa) were obtained at 40 wt.% of TFF, and were 130% and 52% higher than those of S1. The pure matrix has poor bending strength because of its brittleness. Similarly, impact strength and hardness also improved properties at 40 wt.% TFF. This is because of the presence of hard fiber in the matrix. The properties and performance of sample S6 were reduced due to the presence of enormous fibers in the sample, which increased the resin requirement, since the resin weight percent decreased as the fiber increased. This created a shortage of resin to cover the fiber outer surface, which increased the delamination and led to poor bonding. The SEM emphasized the main fracture mechanisms such as delamination, fiber pull-out, voids, and fiber fracture.

The outcome of this study revealed that the prepared sample had superior behavior and environmentally friendly material in all aspects. The results suggest that TFF **Author Contributions:** Conceptualization, S.R. and T.S.; investigation and writing-original preparation, S.R. and F.S.A.; project administration, J.I. All authors have read and agreed to the published version of the manuscript.

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