Development and Characterization of a PLA Biocomposite reinforced with Date Palm Fibers

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ABSTRACT

Despite the promising potential of bio-composites derived from plant fibers due to their ecological and economic benefits, challenges persist in their preparation, restricting their commercial applications. These challenges are primarily associated with developing suitable methods, acquiring appropriate equipment for treating plant fibers, and addressing the time constraints in preparation. This study aims to contribute to the development and characterization of a new biocomposite and biodegradable material based on natural fibers produced through hot compression. The newly developed biocomposite comprises commercial biodegradable poly-lactic acid (PLA) as a matrix and untreated fiber fabric extracted from date palms as reinforcement. The use of untreated fiber fabric has successfully overcome the preparation difficulties. Experimental results on the new biocomposite reveal the strong adhesion between its fibers and the matrix, emphasizing the significant impact of choosing the right manufacturing conditions on the developed mechanical properties.

Keywords-biocomposite; polylactic acid (PLA); Date Palm Fibers (DPFs); mechanical characterization

I. INTRODUCTION

In today's environmentally conscious world, industries like aeronautics, energy, nuclear, and civil engineering are increasingly intrigued by the mechanical capabilities of polymer composites reinforced with natural fibers [1-2]. Notably, cellulose-based fibers such as kenaf [3], flax [4], hemp [5], banana [6], coconut [7], and Posidonia Oceanica [8] are being explored by several researchers. Beyond their commendable mechanical properties, these composites offer the advantage of low density, enhancing structural performance and longevity. Additionally, they stand out as natural, eco-friendly, biodegradable, and recyclable materials. Authors in [9-13] delved into date palm derivatives due to their abundance and diverse fiber types (petiole, eachis, leaflets, fibrillium, bunch, pedicels, spathe, and thorns) [14]. Research on the mechanical characteristics of date palm frond stems was conducted in [15, 16], in which a biocomposite material reinforced with two types of date palm fibers (rachis and leaflets fibers) at varying percentages (4%, 7%, and 10%) was investigated using contact molding for the manufacture of the biocomposites. These cellulose-based fibers are actively used as reinforcements in developing innovative biodegradable and recyclable biocomposite materials. In addition to selecting appropriate fibers, choosing the right matrices is also pivotal in composite manufacturing. These polymers are divided into two classes: thermoplastics and thermosets. Thermoplastics, such as polypropylene (PP) [11, 14] and poly-lactic acid (PLA) [5, 9, 17-19] dominate as matrices for biological fibers, while thermosetting matrices [20] like phenolic, epoxy, and polyester resins are widely employed. Generally, the manufacturing process (method of production, nature of the fibers used, nature of the matrix, etc.) significantly influences the biocomposite performance. There are several manufacturing composite processes for reinforcing with natural fibers, such us injection molding and extrusion, which are instrumental in the plastics industry, involving the careful mixing of raw materials at controlled temperatures. The preparation of natural fibers for composites is a crucial step that requires involving extraction and processing, each with its own associated costs and challenges. The fiber extraction methods vary depending on the nature of the plant, e.g. those for flax, requiring multiple steps such as plucking, retting, scutching, and combing [26]. Special machines, such as those for bamboo and alfa fibers, are used for decortication and are also applicable to banana, sisal, flax, and jute fibers. Date Palm Fibers (DPFs), whether in small pieces or not, necessitate operations like grinding, sieving, cleaning, and drying for reinforcement.

This study presents a contribution to the development of new biocomposite and biodegradable materials at lower cost. Our new biocomposite is composed of biodegradable commercial PLA as matrix and an untreated fiber fabric extracted from date palms as reinforcement. The use of untreated fiber fabric allowed us to overcome the above-mentioned preparation difficulties. The experimental results demonstrate the good adhesion between the fibers and the matrix, and that the right choice of manufacturing conditions has a clear influence on the biocomposite's mechanical properties.

II. MATERIALS AND METHODS

A. Materials

To produce the innovative biocomposite, consisting of PLA as the matrix and DPFs as the reinforcement, we devised and implemented an electromechanical system facilitating its creation through hot compression (Figure 1). The core components of this device include:

- A steel die and punch.
- Dual heating resistors, with one integrated into the die and the other in the punch.
- Twin thermocouples positioned in the die and punch.
- A digital controller tasked with regulating the temperature to the specified set point as needed.

The die, punch, and the upper and lower PLA layers, alongside the DPFs layer are illustrated in Figure 2. The two perforations indicate the positions of the heating resistors and the thermocouples are linked to the temperature controller. The assembly of the die and punch is affixed to an HM-S 200KN-04M4210 traction-compression machine.

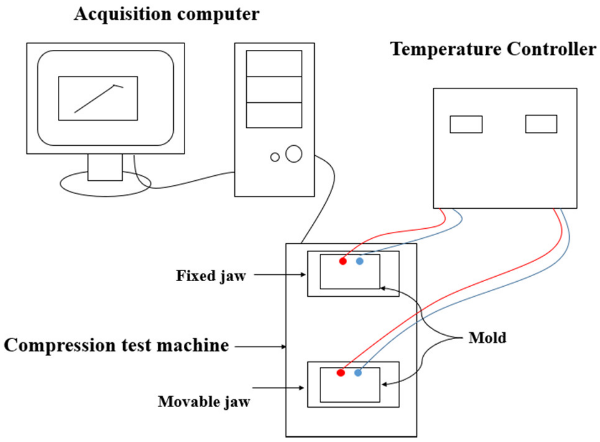

Fig. 1. Schematic diagram of the electromechanical device.

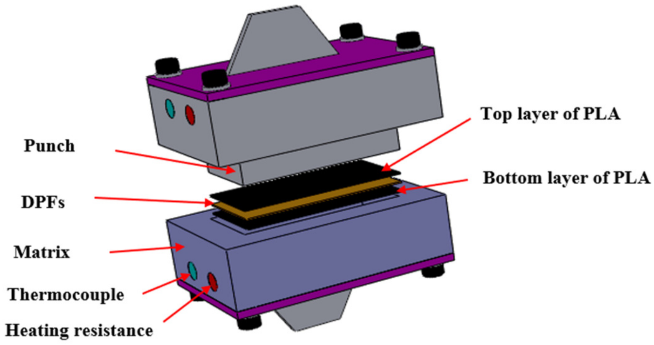

Fig. 2. The experimental device.

B. Methods

Figure 3 outlines the methodology employed in the current research.

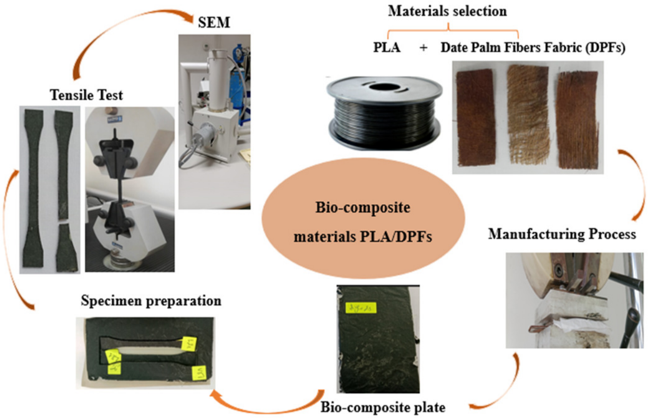

Fig. 3. Manufacturing and testing process.

The characterization process for our novel biocomposite is broadly segmented into three primary stages:

1) Step 1

- Preparation of DPFs fabric measuring 100 mm × 50 mm: Manual cleaning and cutting according to the desired percentage (5% or 10% of the weight of the biocomposite). We note that the fiber fabric has a random structure where some areas may be denser than others, meaning that the distribution of fibers is not uniform over the entire surface. In some places, the fibers may be tighter and closer together, while in others they may be more dispersed. In addition, the yarns that make up the fabric vary in diameter. Some yarns may be thicker, while other, finer yarns may lend additional flexibility and lightness to certain areas of the fiber fabric, as shown in Figure 4.

Fig. 4. Date palm fibers.

- PLA Preparation: The PLA is manually cut into small fragments and it is weighed based on the intended biocomposite composition (PLA/DPFs-5 or PLA/DPFs-10).
- Plates of the new biocomposite through hot compression utilizing the system outlined above and following a predetermined full factorial Design of Experiments (DoE) were generated.

2) Step 2

- Employ laser cutting on PLA/DPFs specimens.
- Conduct a series of tensile tests.

3) Step 3

- Synthesize the findings of this investigation, encompassing the determination of the biocomposite's mechanical properties and the cohesion state of DPFs and PLA.

III. EXPERIMENTS

For the preparation of the novel PLA/DPFs bio-composite, we employed the aforementioned experimental apparatus illustrated in Figures 1-2. This investigation considers five experimental parameters (factors) crucial to the production of the biocomposite: DPFs percentage (5% and 10%), PLA heating temperature T (°C), duration of punch retention in the die during hot compression t (min), cooling duration of the biocomposite plate post-compression t_r (min), and the pressure force required between the die and punch F (daN). These parameters are considered at two levels, as specified in Table I.

To characterize our biocomposite, a series of tensile tests were conducted using a designated apparatus (HM-S 200KN - 04M4210). This machine is equipped with a 10000 daN load cell for recording the applied force, along with pneumatic jaws featuring manual clamping to secure the specimen. The test is displacement-controlled.

TABLE I. EXPERIMENTAL PARAMETERS CONSIDERED FOR THE DEVELOPMENT OF THE NEW BIO-COMPOSITE

Parameter	Description	Level 1	Level 2
T (°C)	PLA heating temperature	160	170
DPF (%)	Percentage of DPFs in the biocomposite	5	10
t (min)	Punch holding time in the die	15	20
t_r (min)	PLA/DPFs plate cooling time	20	30
F (10^3 daN)	Force between die and punch	13	14

The tensile test specimens were laser-cut from the plates of the biocomposite. This selection is justified for two reasons: firstly, laser cutting generally provides precise edges without the need for additional finishing, and secondly, it mitigates issues associated with mechanical cutting. The dimensions of the specimens adhere to NF ISO 527 [27].

IV. RESULTS AND DISCUSSION

In this work, we used PLA as the matrix, whose mechanical properties are: ρ = 850 ± 12 kg/m^3, σ_{Max} = 46 ± 14 MPa, ε= 1.26 ± 0.43 %, and E = 3.32 ± 1.77 GPa. To characterize our PLA/DPF biocomposite and ascertain its mechanical properties, including Young's modulus (E), maximum stress (σ_{Max}), and corresponding strain (ε), a set of tensile tests was conducted on specimens following the ISO 527 standard. These tests facilitated the generation of stress-strain curves depicted in Figures 5 and 6. Figure 5 illustrates the performance of the biocomposite incorporating 5% DPFs (PLA/DPFs-5), while Figure 6 showcases the behavior of the bio-composite comprising 10% DPFs (PLA/DPFs-10).

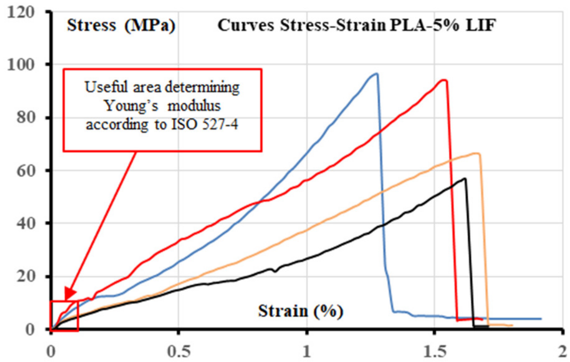

Fig. 5. Tensile curves for samples with 5% natural DPFs.

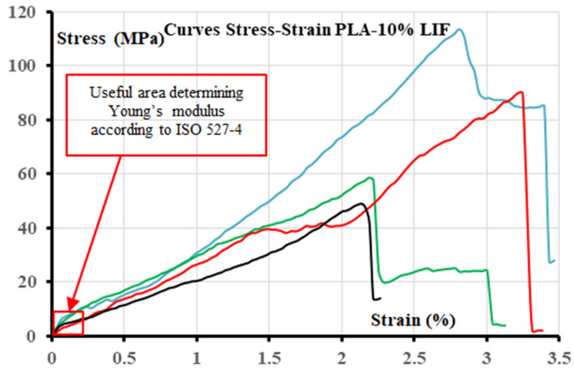

Fig. 6. Tensile curves for samples with 10% natural DPFs.

The tensile behavior of all the specimens is similar and can be broken down into 3 main phases:

The initial phase corresponds to a linear elevation in the applied stress with strain within the [0.05% - 0.25%] strain range, as outlined by ISO 527-4 [28]. This phase corresponds to the elastic region of the stress-strain curve, where the modulus of elasticity can be derived using Hook's law through (1):

$$E = \frac{\Delta\sigma}{\Delta\varepsilon} \quad (1)$$

where E, $\Delta\sigma$, and $\Delta\varepsilon$ represent the modulus of elasticity, the difference in the applied tensile stress between two strain points, and the difference between the two strain points, respectively.

In the second phase, the initial portion of this segment exhibits an almost linear incline, up to the maximum stress value. However, a few peaks within this linear zone are primarily attributed to fiber breaks.

The third and final phase, in which total or degraded stress release is observed, is the phase of total rupture of PLA and DPFs in the bio-composite.

Table II and Figure 7 succinctly present the results of the tensile tests for the two biocomposite types, namely PLA/DPFs-5 and PLA/DPFs-10, as well as standalone PLA. These summaries encompass mean values and standard deviations for the mechanical properties (ρ, σ_{Max}, ε, and E) of the two tensile-stressed biocomposites, comparing them with those of plain PLA. Notably, there is a discernible enhancement in the mechanical property values of PLA/DPFs-5 and PLA/DPFs-10 bio-composites compared to those of plain PLA.

TABLE II. AVERAGE PROPERTIES OF PLA/DPFS BIO-COMPOSITE

Material	ρ (kg/m^3)	σ_{Max} (MPa)	ε (%)	E (GPa)
PLA	850 ± 12	46 ± 14	1.26 ± 0.43	3.32 ± 1.77
PLA-DPFS-5	866 ± 10.5	60 ± 35	1.5 ± 0.2	3.79 ± 2.18
PLA-DPFS-10	871 ± 8.6	75 ± 39	2.8 ± 0.5	3.81 ± 1.91

The results show that the average mechanical properties of PLA/DPFs-10 surpass those of both PLA/DPFs-5 and PLA alone, with substantial increases noted in the majority of cases. Specifically, the average Young's modulus of PLA/DPFs-10 demonstrated a 14.76% augmentation compared to PLA alone and approximately 0.53% when contrasted with PLA/DPFs-5. Similarly, the average maximum strain σ_{Max-10} exhibited a 63.04% increase relative to $\sigma_{Max-PLA}$ and a 25% increase compared to σ_{Max-5}. The average deformation ε_{10} of PLA/DPFs-10 showed a 122.22% increase compared to PLA and an 86.87% increase compared to PLA/DPFs-5. The average density of PLA/DPFs-10 experienced a 2.47% increase compared to PLA and a slight 0.58% increase compared to PLA/DPFs-5. Significant dispersion in the tensile test results was observed for both bio-composites. This is mainly due to:

- The challenging homogeneous PLA distribution between the die and the punch in the produced plates due to the manual placement of PLA fragments in the die. Consequently, achieving uniform distribution of molten PLA on the plates becomes impractical.
- The DPF fabrics in the biocomposite plates are variable in nature. Indeed, the fibers are of random cross-section, length, orientation, and connection to each other (Figure 4).
- The DPFs have not been physically or chemically treated, which may influence the adhesion of the PLA/DPF interface through the presence of fine impurities on the fibers prior to the hot compression operation in the mold.

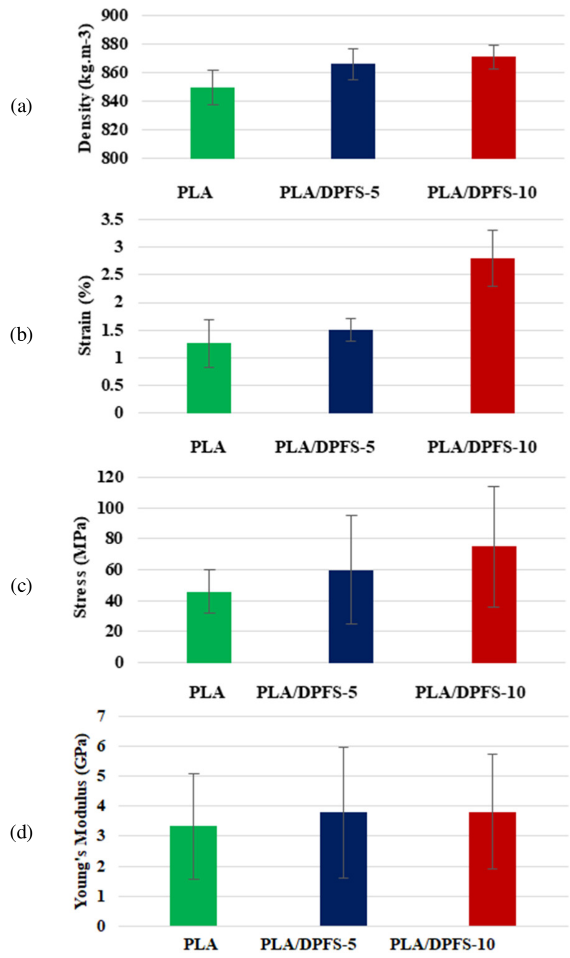

Fig. 7. Variation in bio-composite mechanical properties as a function of DPF percentage.

In order to qualitatively assess the adhesion at the DPF/matrix interface of the new biocomposite, scanning electron microscopy was used to analyze the structure of the new biocomposite. Observations were made on samples cut from damaged specimens. Figure 8 shows an internal view of a plate of the new biocomposite after the hot-pressing operation. It demonstrates the good quality of DPF (10%) and matrix adhesion. This proves that the level of adjustment of the biocomposite's manufacturing parameters is acceptable, as observed on most of the produced plates.

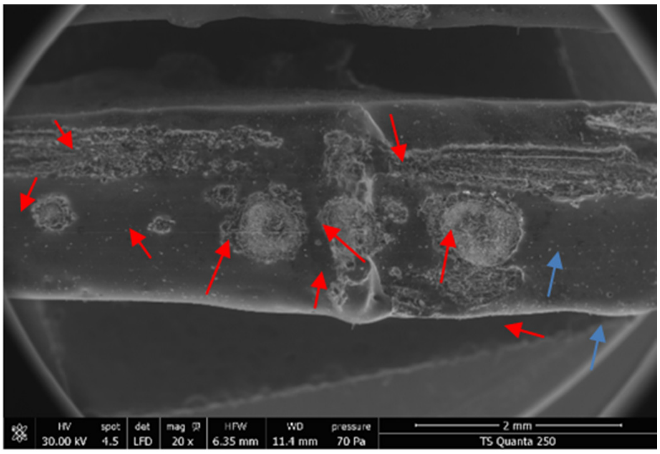

Fig. 8. Adhesion of PLA/DPF-10 interface.

V. CONCLUSION

The primary aim of this study was to propose a cost-effective development and characterization of a novel biocomposite, comprising commercial PLA as the matrix and untreated fiber fabric extracted from date palms as reinforcement. This fabric, abundant in many countries, poses environmental concerns alongside other underutilized waste from date palms and various tree types like olives and almonds. In the initial phase, we engineered a digitally controlled device for producing the new biocomposite through hot compression.

Subsequently, for the characterization of our biocomposite, a series of tensile tests were conducted using a complete factorial experimental design with five factors and two modalities. The outcomes revealed that the average mechanical properties of PLA/DPF-10 surpassed those of PLA/DPF-5 and plain PLA, with significant increases noted in the majority of cases. This underscores the positive influence of augmenting fiber content on the biocomposite's mechanical properties. Furthermore, the results highlighted the high sensitivity of the biocomposite's mechanical properties to slight variations in predetermined manufacturing conditions, such as PLA heating temperature (T), punch retention time in the die during hot compression (t), and the pressure force required between the die and punch of the bio-composite (F). Despite the absence of any physical or chemical treatment, most of the produced biocomposite samples exhibited robust adhesion in all directions between the fiber fabric and the PLA. This was verified by observation using a scanning electron microscope. The current work concerns the development of materials with treated DPFs, 3-point bending tests to characterize the properties of these materials, and the study of the impact of humidity and its effect on the mechanical characteristics of the produced biocomposites.

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