THE INFLUENCE OF USING AGRICULTURE WASTES AS REINFORCING FILLERS ON HYBRID BIOCOMPOSITES PROPERTIES

Nermin M. Aly *[1] and D. E. El.Nashar [2]

[1] *Spinning and Weaving Engineering Dept., Textile Industries Research Division, National Research Centre,33 El Bohouth st., Dokki, Cairo, Egypt,P.O.12622.*

[2] *Polymers and Pigment Dept., Chemical Industries Division, National Research Centre,33 El Bohouth st.,Dokki, Cairo, Egypt,P.O.12622.*

*E-mail: nermin_moh@yahoo.com

ABSTRACT:

Recently, there is a growing interest towards using natural lignocellulosic fibers in reinforcing composites to comply with the global requirements for producing more eco-friendly products. These fibers are offering several desirable properties like renewability, low density, adequate strength, thermal insulation and recyclability. Natural fibers and fillers have proven their success in reinforcement of polymer matrix composites to produce a cost-effective substitute to wood and plastic based products for various industrial applications. Natural fillers can be obtained from several resources such as agricultural wastes which are a good renewable source for lignocellulosic fibers such as date palm leaves, bagasse, rice husk, banana, wood fibers, etc. Their widely usage as fillers in polymeric matrices is owing to their availability, ease of processing and low costs, in addition to improving the mechanical behavior of the produced composites. In this study, hybrid woven sandwich biocomposites samples were fabricated from polyester and flax yarns, using polyester resin as matrix. Agriculture wastes such as date palm leaves and wood flour were used as reinforcing fillers. Chemical modification of lignocellulosic materials was applied to enhance the compatibility between the reinforced materials and the matrix. The hybrid sandwich biocomposites were prepared using grinded date palm leaves and wood flour as filler materials at various weight loading (10, 20 and 30 wt.%). The influence of the fillers content on the performance of the produced biocomposites was examined. Mechanical properties were evaluated throughout impact and flexural tests in both the warp and weft directions of the fabricated biocomposites samples. Nevertheless, physical properties such as water absorption were evaluated to assess their performance. Biocomposites sample's densities were determined for further comparison. Results revealed that, using agriculture waste as fillers have improved the properties of the fabricated hybrid sandwich biocomposites, in which increased the potential of such biocomposites to be used in low cost sustainable structural applications.

Key Words: Date palm leaves, Wood flour, Impact strength, Flexural strength, Water absorption.

1. INTRODUCTION:

The world's nowadays great attention is towards using biodegradable materials in various industrial fields, since the extensive usage of plastic materials had caused severe environmental, economic and social problems. Ecological awareness and sustainability approaches are highly focusing on recycling of products, environmental protection and saving energy. Recently, there were significant achievements in green technology through the development of biocomposites as potential alternatives for wood and plastic based products [1,2]. The usage of natural lignocellulosic fibers as reinforcements in polymer composites for producing cost-effective engineering materials had showed an enormous growth compared to their synthetic counterparts, owing to natural fibers attractive properties like biodegradability, renewability, toxicologically harmless, low density and cost, acoustic properties, acceptable specific strength and modulus and recyclability [3, 4]. Natural fibers such as wood, cotton, flax, jute, hemp, kenaf, coir, sisal, date palm,…etc are available in various forms and are characterized by their high diversity properties [5], they have been exploited as reinforcements or fillers in thermoset and thermoplastic polymer composites due to and have found increasing usage in different industrial applications [4,6,7]. The compatibility between natural fibers and polymer matrices greatly affects on the functional properties of composites, since lignocellulosic fibers have polar hydroxyl groups on their surfaces contributed particularly by cellulose and lignin, which leads to poor interfacial bonding between the fibers and the nonpolar polymer matrix [3]. Many studies have proven that, alkalization is the most effective and simple method for modifying the fibers surface to enhance fibers/matrix adhesion [6,8].

Agricultural wastes utilization in development of biocomposites is currently a matter of interest, due to their benefits to the environment, economy and technology compared to polymer composites reinforced with inorganic fillers. Agricultural and horticultural residues are abundant and can be obtained from various resources such as crops residues, grasses, recycled wood, paper products, etc [2,7,9]. Wood flour is attractive filler for composites applications because of its low cost and wide availability. Date Palm fibers are more effective than industrial materials in terms of their reduced health hazards, processing, renewability and low cost. Several studies have investigated the properties of wood based polymer composites and the potential of using date palm fibers in reinforcing composites and evaluating their performance [8-12]. It was revealed that, natural reinforcing fillers types and content ratios affected the mechanical and physical properties of the composites.

Medupin et al [13], indicated that increasing wood fiber load improved the strength and stiffness of the composites but decreased their impact strength, also water uptake increased with increasing fiber content. Bouafif et al. [14], reported that increasing fiber content in wood based composites increased strength, and decreased energy to break and elongation. Mirmehdi et al [15], found that the flexural and tensile strengths of date palm flour based polyethylene composite was decreased by increasing filler content, while flexural modulus was increased. Recent researches revealed that ,the usage of lignocellulosic fibers in reinforcing polymer composites are more beneficial when used in hybrid composites , that can be produced either by combining various types of fibers, different fabrics structures and configurations in a polymer matrix [2, 4]. Woven fabric composites are finding increasing usage in structural applications because of their inherent advantages and improved mechanical properties, as they provide more balanced properties in the 0◦ and 90◦ directions than unidirectional laminates, besides their ease of handling, ability to produce complex shapes and thicker fiber forms [16-18].

The aim of this work is to study the influence of using agriculture wastes such as date palm leaves and wood flour as reinforcing fillers on the properties of hybrid polyester/flax woven biocomposites. Examinations were performed on the hybrid biocomposites considering mechanical properties such as impact and flexural strengths and physical properties like density and water absorption to assess the performance of the produced hybrid biocomposites for usage as developed value-added products for various applications.

2. EXPERIMENTAL:

2.1 Materials:

Agricultures wastes such as Date palm leaves (DPL) and Wood flour (WF) were used in this work. Date palm leaves were obtained from the date palm trees (*Phoenix dactylifera* L.) in El-Maamoura Botanical Garden, Horticultural Institute in Alexandria. Date palm leaves were extracted from the midribs of fronds with average length 50.6 cm and width 1.3 cm. Wood flour of pine wood was obtained from the sawmill wastes. Polyester/Flax (P/F) woven blended fabrics were produced using polyester fibers in warps and flax fibers in wefts, weaved using warp rib 2/2 and twill 2/2 structures having an areal density of 375 g/m^2 and 377 g/m^2 respectively. Unsaturated polyester resin was used as the matrix with Methyl ethyl ketone peroxide the catalyst and Cobalt Napthanate the accelerator.

2.2. Methods:

Prior to biocomposites processing, the agricultures wastes DPL and WF and the Polyester/Flax woven fabrics were chemically treaded by alkalization.The fibers were soaked in a solution of 1% sodium hydroxide (NaOH), in which DPL were left for 12h, while WF were left for 4h at room temperature with subsequent stirring. The treated fibers were rinsed with water to remove NaOH residues, then neutralized with acetic acid, washed and left to dry in air for 72 hours. Also, woven fabrics were soaked in a solution of 0.5% NaOH for 30 min. at 70°C. to remove natural impurities found in flax,then washed thoroughly with water and neutralized with acetic acid , followed by washing and air drying. After that, the treated DPL were ground into particles using the grinding machine. The treated DPL and WF were screened and sieved using sieve meshes from no.(18-70) to eliminate big and fine fibers. The fibers selected for using in the study have an average length of (1-3mm) and size 210 μm.

2.2.1. Hybrid biocomposites processing:

Date palm leaves and wood flour treated fibers were used at various weight loading (10, 20 and 30 wt.%) as a reinforcing fillers in the hybrid Polyester/Flax biocomposites. Hand lay-up technique was used to fabricate the sandwich biocomposites structure, where woven fabrics were used in the upper and lower skin layers and the fillers form the core layer. The mold used for fabrication of the biocomposites panels was coated using mold releasing agent. Polyester resin, Methyl ethyl ketone peroxide and Cobalt Napthanate were mixed to form the matrix. Fillers content ratio was chosen according to the results of studies applied on these materials [8]. The fabrics were impregnated first with the polyester resin and each filler was added with the specified weight content to the resin, where the filler/resin mixture is added in-between the skin layers and trapped air was squeezed out using a hand roller. The hybrid biocomposites specimens were left to cure at room temperature for 48hours. Table 1 shows the specifications of the produced hybrid sandwich biocomposites.

Table 1. Specifications of the hybrid sandwich biocomposites.

Woven fabric structure	Filler (wt.%)	Biocomposites structure	Thickness (mm)	Weight (Kg/m^2)
Warp rib 2/2	-	P/F	2.9	3.11
Twill 2/2	-	P/F	3.29	3.25
Warp rib 2/2	10	P/F/DPL	3.25	3.7
Twill 2/2	10	P/F/DPL	3.85	4.2
Warp rib 2/2	20	P/F/DPL	4	4.37
Twill 2/2	20	P/F/DPL	4.3	5
Warp rib 2/2	20	P/F/WF	4.61	5.27
Twill 2/2	20	P/F/WF	5.23	6.08
Warp rib 2/2	30	P/F/WF	5.62	6.8
Twill 2/2	30	P/F/WF	6	6.58

2.3. Mechanical and Physical characterization:

The influence of reinforcing filler type and content on the mechanical and physical properties of the produced polyester/flax hybrid biocomposites was examined. Izod impact test was performed according to ASTM D-256 to determine the energy absorbed by the hybrid biocomposites under applying load using CSI-137 Pendulum Impact Tester. The impact strength is calculated according to the following equation:

$$\mathbf{Is = l/(t*w)} \quad (1)$$

Where; Is is the Impact strength (J/m^2), l is the impact load (J), t is the thickness (m) and w is the width (m).
Flexural strength and modulus of the hybrid biocomposites specimens were determined according to ASTM D-790. The test was performed using Universal Testing Machine Galdabini Quasar 50 KN with a cross head speed of 5 mm/min and 2.5 kN load cell. The flexural strength is calculated according to the following equation:

$$\mathbf{Fs = (3pl)/(2bt^2)} \quad (2)$$

Where; Fs is the flexural strength (MPa), p is the maximum load (N), l is the span length (mm), t is the thickness (mm) and b is the width (mm). The flexural modulus is calculated according to the following equation:

$$\mathbf{Ef = l^3m / 4bt^3} \quad (3)$$

Where; Ef is the flexural modulus (GPa), and m is the slope of the tangent to the initial straight-line portion of the load deflection curve.
Density of the hybrid biocomposites specimens was determined according to BS EN-323. Water absorption test was carried out on the hybrid biocomposites specimens according to ASTM D-570, where the biocomposites specimens were submerged in distilled water at room temperature and weighed regularly at different time durations; 2, 12, 24, 48, 72, 96 and 168 hours. Water absorption percentage is calculated according to the following equation:

$$\mathbf{W\%=Wt-Wo/Wo *100} \quad (4)$$

Where; **W** is the water absorption (%), Wt is the wet weight of the specimen at immersion time in water (g) and Wo is the initial weight of the dry specimen (g).
The physical tests and flexural strength test were carried out in the Textile research division laboratories, while the Izod impact test was carried in the Material Testing Laboratory at the Central Unit for Analysis and Scientifically Services at National Research Centre.

3. RESULTS AND DISCUSSION:

3.1. Impact strength:

The impact strength of polyester/flax hybrid biocomposites reinforced with DPL and WF with respect to the warp and weft directions of woven fabrics is presented in figure 1. It was observed that without adding fillers, the warp rib 2/2 biocomposites showed higher impact strength in both directions of fabrics compared to twill 2/2 biocomposites. By adding DPL filler gradually, the impact strength of warp rib 2/2 biocomposites increased by 21.7 % at 10% and by 44.6% at 20% DPL, whereas twill 2/2 biocomposites impact strength decreased by 22.1 % and 27.7%, respectively. This may be related to biocomposites structure which affect on their resistance behaviour during applying load, since twill structure has low intersections which assist in more impregnation of the fibers within the matrix, leading to increasing its stiffness and stress areas which facilitate their failure under load. As well, the higher impact strength value 26.2 J/m was achieved by warp rib2/2 biocomposites in the weft direction at adding 10% DPL compared to the all other specimens. But with increasing filler content up to 20%, the impact strength decreased by 45.6%. Although adding DPL filler to twill 2/2 biocomposites enhanced the impact strength gradually by 30.8 % and 43.4 % with adding 10% and 20% DPL, respectively.

On the other hand, by adding WF filler to the both types of P/F biocomposites, it caused reduction in their impact strength in both directions of fabrics. It decreased in the warp direction for warp rib 2/2 biocomposites by 25.3% and 35%, while for twill 2/2 biocomposites it decreased by 3.9% and 41.6% at adding 20% and 30% WF content, respectively. Where in the weft direction, the impact strength decreased for P/F warp rib 2/2 biocomposites by 40% and 50% and for twill 2/2 biocomposites decreased by 22.7% and 26.6 % at adding 20% and 30% WF, respectively. This loss in their capability of absorbing impact energy can be attributed to the poor interfacial bonding between WF and polyester matrix that caused their brittleness and fracture under load.

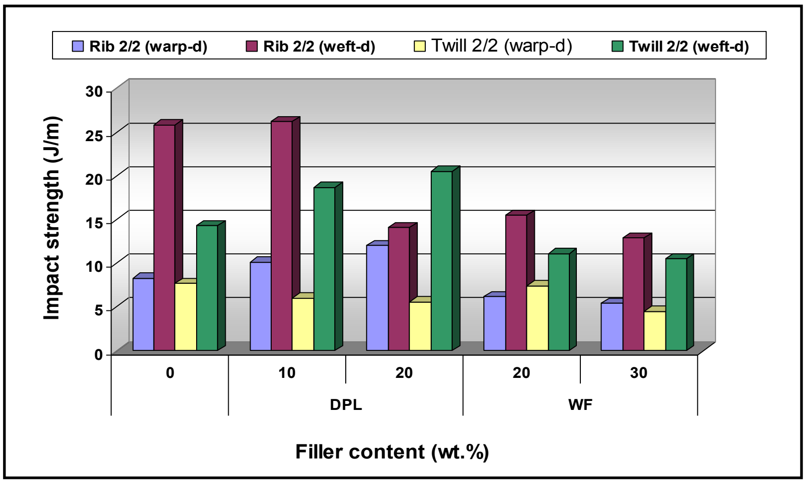


Figure 1. Influence of filler type and content on the impact strength of P/F hybrid biocomposites.

3.2. Flexural strength:

The flexural properties of polyester/flax hybrid biocomposites with respect to the warp and weft directions of skin woven fabrics are presented in figures 2&3. It was observed from figure 2 that, twill 2/2 biocomposites showed higher flexural strength without adding fillers compared to warp rib 2/2 biocomposites. Maximum flexural strength value 113 MPa was exhibited by the warp rib 2/2 biocomposites in the weft direction at adding 20% DPL compared to all biocomposites specimens. The flexural strength of warp rib 2/2 biocomposites in the warp direction increased by 12.84% and 38% at 10% and 20% DPL loading, respectively. Whereas, for twill 2/2 biocomposites it increased by 14% with 10% DPL and decreased by 25% with 20% DPL. This is because twill 2/2 biocomposites reached their maximum strength at 10% DPL and adding more filler leads to decreasing the fibers/matrix interfacial area resulting in agglomeration of filler and increasing the biocomposites stiffness . In the weft direction, the flexural strength values increased with adding DPL up to 20% for both types of biocomposites.

Also, adding WF filler to both types of biocomposites caused reduction in their flexural strength in both directions of fabrics similar to impact properties. The flexural strength of warp rib 2/2 biocomposites decreased by 35.7% and 38.6% and for twill 2/2 biocomposites , it decreased by 48.7% and 52.32% at adding 20% and 30% filler, respectively in the warp direction. In the weft direction, the flexural strength of warp rib biocomposites decreased by 28.6% and 33.45%, whereas for twill 2/2 biocomposites, it decreased by 34.8% and 42.2 % at adding 20% and 30% filler, respectively due to poor wettability of and interfacial bonding between WF and polyester matrix.

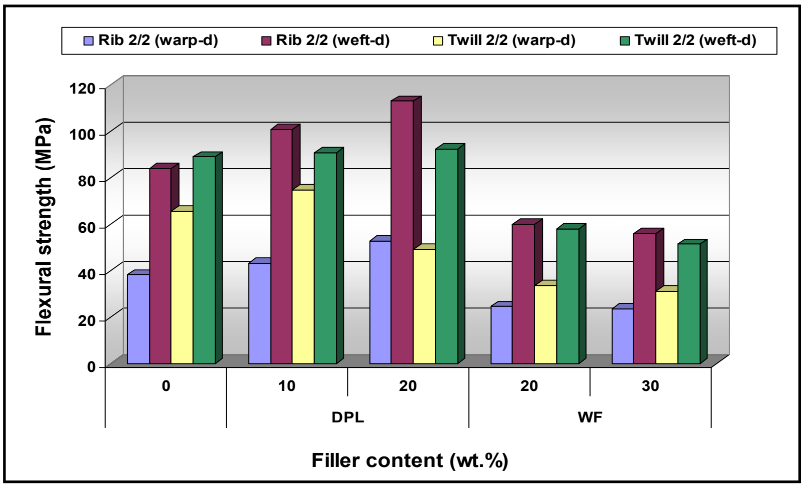


Figure 2. Influence of filler type and content on the flexural strength of P/F hybrid biocomposites.

Flexural modulus of polyester/flax hybrid biocomposites is shown in figure 3. It was observed that, the higher flexural modulus values were recorded in the weft direction compared to that in the warp direction. Maximum flexural modulus value 3.6 GPa was exhibited by warp rib 2/2 biocomposites at adding 20% DPL compared to all specimens. Flexural modulus of warp rib 2/2 biocomposites improved with adding DPL up to 20% in both directions of fabrics. It was noted that, twill 2/2 biocomposites showed its highest flexural modulus in the warp direction with 10% DPL, as it increased by 34.9% and reduced with adding 20% DPL. Further, it showed an increasing trend in the weft direction as it increased by 14.4 % and 28.7% at 10% and 20% DPL respectively. Even though adding WF filler to the warp rib 2/2 biocomposites, it caused improvement in their flexural modulus up to 30% in both directions of fabrics. This can be related to WF fibers high modulus compared to DPL. The flexural modulus increased by 30.14% and 45.8% in the warp direction, and increased by 15.4 % and 48.6 in the weft direction at adding 20% and 30% filler, respectively. Whereas the twill 2/25 biocomposites showed slight increase in their flexural modulus with adding 20% WF. But at adding 30% WF, the flexural modulus decreased due to agglomeration problem of WF which cause poor interfacial adhesion strength between the filler and polyester .

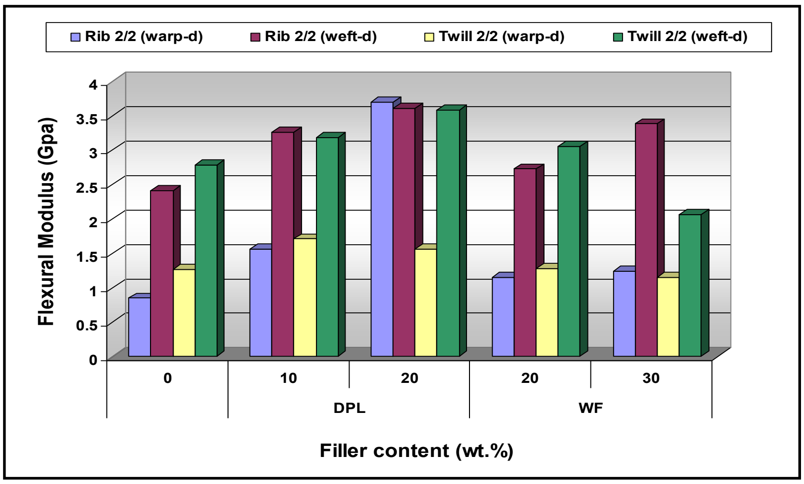


Figure 3. Influence of filler type and content on the flexural modulus of P/F hybrid biocomposites.

3.3. Density:

Figure 4 shows density values of the polyester/flax hybrid biocomposites specimens. It was observed that, the density of the hybrid biocomposites increased gradually with increasing the content of both fillers. The hybrid biocomposites reinforced using DPL showed lower densities compared to those reinforced with WF. This could be attributed to the low density of date palm leaves compared to wood fiber density. Also, warp rib 2/2 biocomposites recorded higher density values compared to twill 2/2 biocomposites. Highest density value 1.25 g/cm^3 was found in the warp rib 2/2 biocomposite reinforced with 30% WF content.

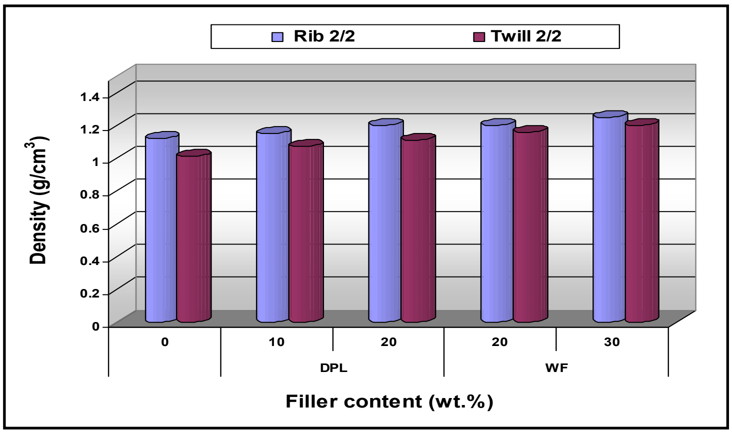
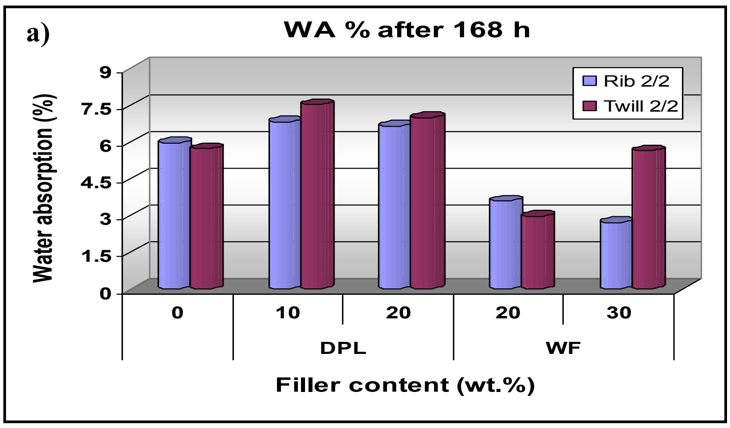


Figure 4. Influence of filler type and content on the density of P/F hybrid biocomposites.

3.4. Water absorption:

Figure 5a) shows water absorption behaviour of polyester/flax hybrid biocomposites specimens after immersion in water for 168 hours. It was indicated that, the warp rib 2/2 biocomposites with 30% WF content showed the lowest water absorption rate of 2.7% after 168 h, followed by the twill 2/2 biocomposites with 2.9% at adding 20% WF content. While the highest overall water absorption 7.5% was observed in the twill 2/2 biocomposites at adding 10% DPL. This could be attributed to high stiffness of the biocomposites specimen and to the warp rib2/2 fabric weaving structure which is characterized by more yarns intersections compared to twill structure that accordingly leads to less porosity and limited water penetration. Figure 5b) shows water absorption rate of warp rib 2/2 biocomposites at adding 30% WF during the whole experiment. It was found that, the specimen highest water uptake rate was in the first 24 hours, then the rate decreased considerably until 72 h and almost stopped beyond 96 h. Also the twill 2/2 biocomposites with 20% WF behaved in a similar manner.



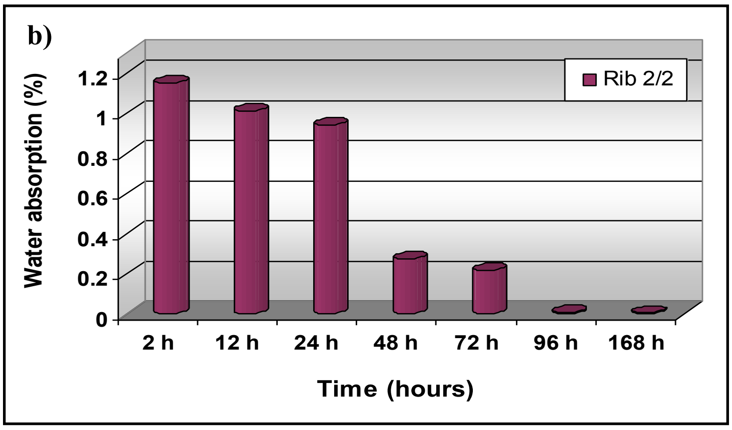


Figure 5. a) Influence of filler type and content on water absorption of P/F hybrid biocomposites after 168h, and b) water absorption rate of warp rib 2/2 biocomposites with adding 30% WF**.**

4. CONCLUSION:

In this study, the mechanical and physical properties of polyester/flax woven biocomposites reinforced using date palm leaves and wood flour at different weight loading were investigated. The results revealed that, woven fabric materials and structure affected on the performance of the produced hybrid biocomposites, in addition to the filler content ratio. The hybrid biocomposites achieved higher mechanical properties in the weft direction of fabrics compared to those in the warp direction. The impact and flexural strengths of warp rib 2/2 biocomposites improved in the warp direction with using 20% DPL due to the good interfacial bonding between DPL and polyester resin. While in the weft direction it reached its maximum strength at 10% filler loading. Twill 2/2 biocomposites showed better behaviour and higher impact and flexural strengths in the weft direction, while in the warp direction only the flexural strength enhanced with using 10% DPL. Adding WF filler to both types of biocomposites caused reduction in their impact and flexural strengths in both directions of fabrics. Although, it caused improvement in the flexural modulus of warp rib 2/2 biocomposites up to 30%. The lowest water absorption was observed in warp rib 2/2 biocomposites with using 30% WF after 168 h immersion in water. Thus, utilization of agriculture wastes in reinforcing hybrid biocomposites could be a base for developed value-added green products with enhanced properties for various industrial applications.

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