Physical properties of coir and pineapple leaf fibre reinforced polylactic acid hybrid composites

R Siakeng¹, M Jawaid¹,², H Ariffin³ and S M Sapuan¹

¹Laboratory of Biocomposite Technology, Institute of Tropical Forestry and Forest Products (INTROP), Universiti Putra Malaysia, 43400 Serdang, Selangor, Malaysia
²Department of Chemical Engineering, College of Engineering, King Saud University, Riyadh, Saudi Arabia
³Laboratory of Biopolymer and Derivatives, Institute of Tropical Forestry and Forest Products (INTROP), Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia

Email: ramengmawii8@gmail.com

Abstract. This study examined the physical behaviour of Coir fibres (CF) /Pineapple leaf fibres (PALF) /Poly lactic acid (PLA) composites. In this research, coir and PALF reinforced PLA hybrid composites were fabricated by hand lay-up process and hot press. The aim of this work is to do comparative study on density, water absorption (WA) and thickness swelling (TS) of untreated CF/PALF reinforced PLA composites and hybrid composites. The effect of different fibre ratios in hybridization on density, WA and TS of CF/PALF hybrid composites were also analyzed and C7P3 showed highest density while P30 had lowest. The results indicated that the density varies on different fibre ratio. WA and TS of CF/PALF composites and hybrid composites vary with fibres ratio and soaking duration. WA and TS of untreated CF/PALF hybrid composites were increased by increasing coir fibre ratio so, C30 showed highest WA and TS whereas P30 and C1P1 showed least WA and TS respectively apart from neat PLA.

1. Introduction
Recently, natural materials have attracted substantial interest due to the rising need for lowering the environmental load and to ensure sustainability [1, 2]. This has also led to a considerable change in the research direction of fibre reinforced polymer composites; which is a biodegradable composite composed of a biodegradable polymer and natural cellulosic materials which are expected to be environmentally friendly [3-6]. Polylactic acid (PLA), made from renewable raw materials has been considered to be a type of commercially available thermoplastic polymer and fully biodegradable [7, 8]. PLA is now one of the most promising biodegradable polymers for industrial plastic applications, as an alternative to conventional synthetic polymers due to its high mechanical properties and good processability [8, 9]. Nonetheless, it has some limitations such as brittleness, low impact strength, and low thermal resistance [8, 10]. In order to make them suitable in various applications, the mechanical properties of PLA needs to be enhanced by using reinforcements such as natural fibres [1, 11]. Natural fibres such as coconut coir, jute, hemp, kenaf, pineapple leaf fibres can be combined with a polymer.
matrix to produce biodegradable composites with specific properties comparable to synthetic fibre based polymer composites [12-18].

Coconut coir is a multipurpose lignocellulosic fibre extracted from the husk of coconut and is one of the stiffest among the tough natural fibres and draw large attention due to their potential to reinforce polymer and bulk density to yield cost-effective composite products [19]. Coir fibres have lower cellulose (36–43%) and hemicellulose (15.17%) contents, higher lignin content (32.25%) and microfibrillar angle (30–45°) higher than other natural fibres [20] which results in their relatively low tensile strength and good impact as well as highest elongation at break among natural fibres [2]. Coir drawn much attention due to their good thermal conductivity and biodegradability properties [13]. Likewise, PALF also has the potential for use as reinforcing filler biocomposites with gratifying physical and mechanical properties [21]. PALF is a smooth thin yet strong natural fibre with high cellulose content (70-82 %), lignin (5-12 %) and ash (1.1 %) [22].

Natural fibre reinforced polymer composites are sensitive to moisture and humidity. Water absorption affects the mechanical strength and fibre-matrix interfacial bonds leading towards poor stress transfer efficiencies from matrix to fibres [17]. Study of water absorption in natural fibre composite is necessary for various applications such as outdoor component, packaging and building industry [23]. In natural fibre reinforced composites, fibre absorb water due to many reason such as temperature, fibre loading and orientation, fibre characteristics, area of exposed surfaces, diffusivity, and surface protection [24]. Mechanism of water absorption in fibre composites is based on three ways; first method is diffusion method, water molecules flow into micro-cracks of polymer chains. Second method is capillary flow; water flow along with the interface if interfacial bonding of fibre and matrix is weak. Third is moisture content; storage of water in micro-cracks present in both polymer and natural fibre [25]. Various research [23, 26, 27] have been conducted on the water absorption and thickness swelling of different types natural fibre composites and discussed the problem associated with water absorption in composites. In this research water absorption and thickness swelling properties of Coir/PALF/PLA composites have been studied on various fibre ratios and time span. The objective of this work, fibre mixing ratio of Coir/PALF/PLA composite are optimised by water absorption and thickness swelling testing, it will be helpful for further study of mechanical, thermal and biodegradability properties of the same biocomposites.

2. Experimental procedures

2.1. Materials
Polylactic acid (PLA) pellets with a molecular weight \(M_w\) of approximately 74,000 g/mol, density of 1.25 g/mol, glass transition temperature \(T_g\) of 55 to 60°C and melting point \(T_m\) of 160 to 180°C was purchased from TT Biotechnologies Sdn. Bhd., Malaysia. Coir fibres \(\text{(Cocos nucifera)}\) were harvested in west Malaysia while pineapple leaf fibres (PALF) \(\text{(Ananas comosus)}\) were harvested in India and was supplied by Innovative Pultrusion Sdn Bhd., Malaysia.

2.2. Preparation of composites
Coir and PALF used as reinforcements in fabrication of composites were chopped into short fibres (1-2 mm) by using ring flakers machine; fibres were maintained at 6-8% moisture content. Coir and PALF composites were fabricated by using 15x15x3 mm stainless steel plate mould. The mould was placed into hydraulic pressure hot press at 180° temperature and was removed from the press after 15 minutes pre-heat and 5 minutes press kept for cooling in cold press for 3 minutes, and then samples were cut for testing according to ASTM standard. The formulations of hybrid composites were described in table 1.
Table 1. Density and formulation of Coir/ PALF/PLA hybrid composites.

| Composite Code | PLA (Weight %) | Coir (Weight %) | PALF (Weight %) | Density (g/cm$^3$) |
|----------------|----------------|----------------|-----------------|---------------------|
| Pure PLA       | 100            | —              | —               | 1.2                 |
| C-30           | 70             | 30             | —               | 1.13                |
| P-30           | 70             | —              | 30              | 1.02                |
| C1P1           | 70             | 15             | 15              | 1.07                |
| C3P7           | 70             | 9              | 21              | 1.05                |
| C7P3           | 70             | 21             | 9               | 1.09                |

2.3. Composite density determination
Density was measured by using ASTM D 1895 standard. The density of the samples was calculated by using following equation (1):

$$\rho = \frac{m}{v}$$  \hspace{1cm} (1)

where, $m$ represents mass of fibre/matrix and $v$ represents volume of fibre/matrix.

2.4. Water absorption test
Prepared samples were immersed into distilled water at room temperature. Water absorption was calculated by the equation (2) for various period of time. All the samples were weighed before and after soaking and recorded carefully. At the three intervals, water absorption was examined such as 3 day, 5 days and 7 days. After taking out samples from water, all the samples were wipe dry thoroughly to remove excess amount of water on the surfaces. The percentage equilibrium water absorption was calculated as an average value of several measurements. The percentage of water absorption was calculated from equation (2) using ASTM D570 [28].

$$\text{Water absorption (\%) } = \frac{W_n - W_d}{W_d} \times 100$$  \hspace{1cm} (2)

where, $W_n$ is the weight of composites samples after immersion and $W_d$ is the weight of the composite samples before immersion.

2.5. Thickness swelling test
Six samples of pure PLA and each different fibre ratio combination composites were prepared for the experiment of thickness swelling. Thickness swelling was calculated according to ASTM D570 by using Equation (3) [28]. The thickness swelling experiment was conducted by measuring the swelling of the each sample. Measurement of the composites was taken before and after the soaking. Samples were measured three times, first measurement was taken on third day of immersion second was fifth days of immersion and last was seven days of immersion.

$$\text{Thickness Swelling (\%) } = \frac{T_1 - T_0}{T_0} \times 100$$  \hspace{1cm} (3)

where $T_1$ is the thickness after soaking and $T_0$ is the thickness before soaking.
3. Result and discussion

3.1. Density
The experimental densities of untreated coir/PALF reinforced PLA composites and hybrid composites were calculated shown in table 1. The density of P30 composites is lowest (1.02 g/cm³) among all composites. However C30 composites showed highest experimental value (1.13 g/cm³). The experimental density of C7P3 is highest among the hybrid composites which is 1.09 g/cm³ which is lower than the coir/PLA composite and higher than PALF/PLA composite. The differences among composites are mainly due to manufacturing process and fibre ratio or loading. In some case, composites have void space due to air trap inside composites [29].

3.2. Water absorption
Water absorption test was conducted to study the amount of water absorbed in a definite period of time. Generally, the natural fibre composites absorb moisture due to several reasons such as fibre fraction, voids, viscosity of matrix, humidity and temperature [29]. As shown in the Error! Reference source not found. all untreated composites showed high WA than neat PLA sample. Among the all composites, P30 composites absorb lowest moisture contents, while C30 composites showed highest WA which showed 10% WA increment. This study indicated the presence of void content and porosity on the surface of untreated composites. Due to the presence of voids and pores on the surface of composite, the weight of composite increases by trapping the water inside the voids [29]. On the other hand, the WA for the pure neat PLA was lowest among all. In this result, it is shown that all hybrid composites showed quite similar pattern of WA rate having the pattern of increased WA with increasing coir fibres ratio.

There are many factors such as porosity, void content, lumen size, and fibre-matrix adhesion to effect water absorption behaviour of the composites besides the hydroxyl groups present in cellulose and hemicelluloses of natural fibres which provides easy access to moisture in to composites [30]. The immersed composites into the water absorb water molecules through capillary action and fills the voids and cracks in the composites [28, 31]. The higher WA rates in coir reinforced composites compared to PALF could be due to the higher porosity and hollowness of the coir fibres.

![Figure 1](Image)

**Figure 1.** Water absorption of untreated coir/PALF/PLA composites.

3.3. Thickness swelling
Thickness swelling test was conducted to study the affect of water molecules on dimension stability of coir/PALF reinforced PLA composites. The TS is the exposure of the natural fibre on the surface of composite. The hydrophilic nature of lignocellulosic materials and the mechanism called capillary action caused the intake of moisture when the samples were soaked in water which leads to the swelling of fibres and thus increase the dimension of composite [29]. Figure 2 shows that neat PLA
sample showed high dimensional stability, however, composites and hybrid composites revealed higher thickness swelling. Among the all composites, C1P1 hybrid composite showed highest dimension stability and C30 composites showed highest thickness swelling approximately 8%.

Higher TS indicated the presence of void on the surface or high porosity in the composites. It is responsible for the changes in the dimension of lignocellulosic fibre-based composites, particularly in the thickness, and the linear expansion due to reversible and irreversible swelling of the composites [31]. C30 composites have high content of coir fibres which is a hollow and porous lignocellulosic fibre [19], which may be the cause of thickness swelling.

![Figure 2. Thickness swelling of untreated coir/PALF/PLA composites.](image)

4. Conclusion
In this work, density, water absorption and thickness swelling were examined. 30% coir fibre reinforced PLA composite showed highest water absorption and thickness swelling than all other tested composites and hybrid composites. Inclination of fibre composites were obvious, higher coir fibre loading showed higher moisture content and swelling in the composites. Neat PLA showed lowest water absorption which indicates presence of less void contents. Thickness swelling is directly related to the water absorption, therefore, the trends of water absorption and thickness swelling of all composites are very similar. High water absorption and thickness swelling affect the interfacial bonding between fibre and matrix. In this study we found that water absorption and thickness swelling did not much affect the physical properties of composites due to very less percentage of water absorption and thickness swelling observed. However, natural fibres are hydrophilic due to the presence of hydroxyl groups from cellulose and lignin. So, surface modification of these fibres would improve the fibre–matrix bonding and hence gives better composites products. The results of the present study show that a useful composite with good strength could be successfully developed using coir fibres as a reinforcing agent for the polylactic acid matrix.

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