The effect of fibre loading on the physical and mechanical properties of Pineapple Leaf Fibre-reinforced biodegradable composites

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Abstract. Recently, research on the development of biodegradable material were performed. However, biodegradable materials have a crucial weakness in terms of mechanical properties, but it can be improved by adding reinforcement material as filler. In this study, Pineapple Leaf Fibre (PALF) was chosen as filler due to its many advantages, such as low cost, variety and redundant sources and bio-degradable. The objective of this research is to investigate the effect of PALF fibre loading to the corn starch composites in terms of physical, mechanical and morphological properties. The samples were prepared at different fibre loadings: 5 wt%, to 70 wt %, and were fabricated by using the hot press machine. The result showed that physical properties, such as density, water absorption and moisture content, were affected by different fibre loadings. Mechanical properties of the composite samples showed that PALF with 30 wt% gave the optimum tensile strength and modulus. Morphological analysis, composite samples of 30 wt% PALF fibre loading had fewer gaps between matrix and fibre. It can be concluded that 30 wt% of fibre loading is the optimum fibre loading for 3cm length PALF as compared to other composition ratios.

1. Introduction

Nowadays, the world is facing many problems regarding environmental issues. Some of the issues include harmful industrial materials that can affect human health, disposal of non-biodegradable materials such as plastic and global warming caused by the greenhouse effect. To overcome these arising problems, researchers from all over the world are considering the production of environmental friendly materials or also known as green materials. One of the solutions that were recently highlighted by research is replacing synthetic fibre composites with natural fibre composites because natural fibre has many advantages as compared to synthetic fibre. The advantages of natural fibre are [1] low in density [1–4], high specific strength and stiffness [5,6], renewable resource, can be produced at a cost that is lower than synthetic fibre, low hazard manufacturing processes, low emission of toxic
fumes when heated and during incineration, and less abrasive damage to processing equipment as compared to synthetic fibre composites. There are many bio-composite products that are commercialized from fibre-based plants, such as kenaf, jute, roselle, sugar palm fibre and banana pseudo stems.

One of the natural fibres that has good properties is pineapple leaf fibre (PALF) [5, 7, 8]. PALF is a natural fibre that has potential to be used as reinforcing materials in green composite products. It is very marketable in Malaysia due to the fact that it is one of the largest plantations in Malaysia that has over 130 distinct cultivated pineapple species. Currently, the country’s pineapple industry is mainly focused on the fruit and related foodstuffs, while the pineapple leaves are treated as agricultural wastes, which are either composted or burned by farmers. This leads to the wastage of a very potential source of good natural fibre.

Research found that the PALF properties vary according to the plant types, geographical regions, plant age, and weather conditions [9]. Non-treated and treated PALF of various lengths and fibre loadings are used to reinforce matrices, such as polypropylene, polyethylene, polycarbonate and polyester [2].

A new direction in biodegradable polymer development from renewable resources is provided by the development of synthetic polymers by using monomers from natural resources. One of the most promising polymers in this matter is PLAF, since it is made from agricultural products and is readily biodegradable [3][4]. In literature, for the past few years, the application of starch resources in non-food applications had experienced a significant advancement. Starch has many benefits, such as low cost, wide resources, and total compostability [10], [11]. Like many other polymers, starch uses the conventional polymer processing techniques that produce different end-use forms, for example, extruded, moulded, and thermoformed. However, starch-based materials are known to have limitations, like poor processing ability and properties, such as weak mechanical properties, poor long-term stability, and high-water sensitivity.

However, these starch weaknesses can be improved by adding a plasticiser. At elevated temperatures and when the shear is present, starch will exhibit thermoplastic properties which are close to synthetic polymers. Therefore, for starch processing, it is possible to use procedures that are developed for synthetic polymers. Corn starch is one of the potential biodegradable plastics to be used widely in the future.

An extensive research was done on corn starch properties for application in packaging as bioplastic material [5]. The study emphasised on the mechanical behaviour of corn starch and corn starch plastic stability towards water resistance. This research discusses the Pineapple Leaf fibre reinforced cornstarch composites (PLCS) mechanical and physical behaviours. In this research, PALF was treated by using alkali treatment to obtain a clean fibre surface. The mechanical and physical behaviours are thoroughly discussed.

2. Samples Preparation
PALF was purchased in Johor, Malaysia. The raw PALF was treated with alkali solution to remove impurities on its surface and improve the fibre surface properties. It was first soaked in 5% of NAOH solution for 4 h and then cleaned by using distilled water. The fibres were left to dry at room temperature for about 24 h. They were manually cut into 3 cm length as shown in Figure 1.

**Figure 1.** Fibre preparation as a filler for cornstarch biodegradable composites.

The matrix used in this research was a mixture of corn starch and glycerol. Glycerol acted as a hardener that helped to reduce the wetting ability of corn starch to ensure that the composite sample
was not affected by moisture. Firstly, the corn starch weighed 210 g, while the glycerol was 90 g. To ensure a uniform mixture the corn starch and glycerol were mixed with a mixer. The sample was fabricated by using the hand lay-up method, and was preheated under a hot press machine for about 15 min. The temperature and pressure settings for the pressing process were 165°C and 350 kPa, respectively. After the pre-heat procedure was completed, the mould was pressed for another 15 min and cooled for about 15 min, as shown in Figure 2.

3. Methods
3.1 Density
The density of the PALF reinforced corn starch biodegradable plastic composites was determined by using an electronic densimeter. Six samples for each fibre loading of PALF-reinforced corn starch composites were prepared for density measurement. Prior to being immersed in water, the weights of all samples (m) were recorded. The volume (V) of samples was recorded by looking at the amount of water before and after sample immersion. This value was used to determine the density of PALF reinforced corn starch composites.

\[
\rho = \frac{m}{V}
\]  

(1)

3.2 Moisture Content
Six samples (10 x 10 x 3mm) were prepared for moisture evaluation. All samples were heated in an oven at 105°C. The weight of the samples before and after heating \((M_i)\) and \((M_f)\) were measured as the values were used to calculate the moisture content through Equation 2:

\[
\left( M_i - \frac{M_f}{M_i} \right) \times 100
\]  

(2)

3.3 Water Absorption
Six samples (10 mm x 10 mm x 3 mm) were dried in an air circulating oven at 105°C±2 for 24 h to remove moisture. Then the samples were immersed in water at room temperature (23±1°C) for 30 min and 2h. The samples were weighed before immersion \((W_i)\), and after immersion \((W_f)\). The water absorption of the samples was calculated by using Equation 3.

\[
\left( W_i - \frac{W_f}{W_i} \right) \times 100
\]  

(3)

3.4 Mechanical Testing
3.4.1 Flexural Test
Flexural tests were conducted by using a three-point bending setup according to the ASTM D7264 standard (ASTM, 2007). At least five samples were tested by the flexural method. The dimension of the samples was 140 mm (l) x 13 mm (w) x 3mm (t), and they were tested by using a Universal Testing Machine called Instron. Results were determined and recorded in Bluehill 2 Software. The load speed applied for this test was 1mm/min. The samples were placed on a gauge with the head adjusted to touch the surface of the samples and the load was applied onto the samples, as shown in
3.5 Morphological Analysis

For this research, a morphological study was done in detail on the fractured surface of the tensile test samples. This was performed by using a Scanning Electron Microscope (SEM) as shown in Figure 4. The selected samples contained 10 wt%, 30 wt% and 70 wt% of fibre loading. The samples were coated with platinum to offer a good electrical conductivity and obtain results as the resolution was not significantly affected.

4. Results and Discussion

4.1 Density

Density of a material is defined as its mass per unit volume. It is a term used to show the relation of the composite weight to its size. Density is important as it affects the sample weight.

From Figure 5, the density of the composite seems to increase with increase in fibre loading. This means that the higher the weight percentage of fibre loading, the higher the density of the PALF-reinforced corn starch composites. At the lowest fibre loading of 5 wt%, the density was only 1.2924 g/cm$^3$. The increase was from 1.2924 g/cm$^3$ at 5 wt% of fibre loading to 1.396 g/cm$^3$ at 70 wt% of fibre loading. The increased density was about 7%, which can be concluded that it was not significantly different and the composite samples still gave the lightweight effect as compared to neat plastic.

The higher the fibre loading, the higher is the density of composites. This will affect the composite weights as density is directly related to the weight of a substance, or in this case, the composite.
4.2 Moisture Content and Water Absorption

Even though natural fibre has many advantages related to its usage as a reinforcement material, it has some weaknesses. Natural fibre has drawback in obtaining a good adhesion between fibre and matrix due to its hydrophilic behaviour. Therefore, the resulting natural fibre has high moisture content and high water absorption properties. This will weaken the composite product application [12]. Nevertheless, this disadvantage was overcome by performing surface treatment on the natural fibre, or in this research the PALF. Past studies showed that treated natural fibre has better hydrophilic adhesion as compared to untreated fibre [13]. In natural fibre-reinforced composites, water is transported through a multiple natural plant mechanism. The penetration of water molecules is enabled by the micro-crack in the polymer matrix. Other than that, water may also be transported through the fibre and matrix interface, which will result in unwanted changes or deformation of mechanical properties, for example, swelling, plasticising and degradation.

Figure 6 illustrates the moisture content of the PALF-reinforced composite increase directly with the fibre loading wt%. At 5 wt% of fibre loading the moisture content is 0.045, which is the lowest reading. Meanwhile the highest moisture reading is at 70 wt% fibre loading with a moisture content of 0.089. These readings showed that the moisture content of a composite is affected by its fibre loading. The higher the presence of fibre in the composition, the higher is the moisture content.

As a conclusion, the moisture content of the composite is affected by its fibre loading. From this study, it was proven that higher fibre loading lead to higher moisture content. High moisture content affects the interface between the fibre and matrix.

The water absorption testing was divided according to its soaking time. The sample soaking time was 30 min and 2 h. From Figure 7, it can be observed that water absorption showed different results, depending on its soaking time. The composite that was soaked for 2 h absorbed more water as compared to the sample that was soaked for only 30 min. Furthermore, the fibre loading also affected the water absorption. Higher fibre loading sample has larger water absorption. This can be seen through the comparison of samples for both soaking time of each fibre loading. For example, the sample with 30 wt% fibre loading had different water absorption reactions for both soaking time. Water absorption when the sample was soaked for 2 h was larger as compared to the sample that was soaked for 30min.

By comparing the fibre loadings, higher percentage of fibre loading have higher water absorption. In Figure 7, fibre loading of 5 wt% has lower water absorption as compared to 70 wt% fibre loading.
To conclude, it can be observed that water absorption of the composite can be affected by the soaking time and fibre loading. The longer the soaking time, the higher is the water absorption of composites. Similar to moisture content, the bigger the percentage of fibre loading, the higher is the water absorption. High water absorption will cause composite degradation which will lead to composite disfunction in its application.

4.3 **Flexural Properties**

Flexural testing is a test that is performed on a material to determine the properties of flexural or bending of material. The test is conducted by placing the sample between supports and putting the load by using a third point which is known as the three-point bend. Flexural test gives information on the elasticity modulus value in bending and flexural stress of the tested material.

From Figure 8, the PALF-reinforced corn starch flexural stress and the modulus of elasticity at 5 wt% fibre loading is 1.95 MPa and 72.409 MPa, respectively. Starting from 5 wt% fibre loading, both flexural test and sample modulus show increment until the fibre loading finally reached 30 wt%, whereby the highest reading was recorded. The readings were 10.59 MPa for flexural stress and 532.41 MPa for elasticity. However, at 30 wt% onwards, the flexural stress and composite modulus started to decrease until 70 wt% of fibre loading.

The adhesion between the PALF and corn starch can be the reason to the changes in reading. At the range of 5 wt% until 30 wt% of fibre loading, the stress was increased because the adhesion between fibre and matrix was strong and completely bound the fibre. Meanwhile for the 40 wt% until 70 wt%
fibre loadings, the value for both properties started to drop. This might be due to the increase in fibre loading of the composite composition. The high percentage of fibre loading affected the adhesion bond between corn starch and PALF. This was because the matrix was unable to cover all fibres due to the decrease in percentage of corn starch used. The corn starch was unable to disperse completely between the matrix, resulting in a weak binding between the PALF and corn starch[7].

As a conclusion, the percentage of fibre loading effects the flexural properties of the PALF-reinforced corn starch composite. This can be proven by the result that was previously explained. The sample flexural stress and elasticity modulus were optimum at 30 wt% as it had the highest value of both properties. However, when the fibre loading started to overload, the matrix was unable to disperse as the fibre loading was higher than the matrix itself. This resulted in some problems like wetting and void.

By using SEM, the morphological analysis on the PALF-reinforced corn starch composite was done. This test was performed to analyse the composite particle characteristic after it underwent the tensile test. For this analysis, three samples from the 10 wt%, 30 wt% and 70 wt% was chosen. 10 wt% was chosen as the initial sample as it contained appropriate amount of fibre to be analysed as compared to the 5 wt% fibre loading. Meanwhile the 30 wt% was chosen based on the tensile test result, in which at this fibre loading, the PALF-reinforced corn starch material had the highest Ultimate Tensile Strength (UTS); thus, resulted as the optimum sample as compared to the other fibre loadings. The 70 wt% fibre loading was chosen as the final sample due to the fact that it showed the lowest UTS reading as compared to other fibre loadings, after the maximum value of UTS at 30 wt% of fibre loading.

From Figure 9 (a), it can be seen that there is a clear gap between the PALF and corn starch. A separation between the matrix of composites can also be observed. These defects were due to the low weight percentage of the fibre in the composite, and thus affecting the binding strength of the composites, resulting in a low tensile strength value with the sample of 10 wt% fibre loading. Figure 9 shows the internal condition of the optimum fibre loading sample, which was at 30 wt%. At this fibre loading, the adhesion between the fibre and matrix showed no gap. It could be observed that the PALF was completely covered by the corn starch. Even though there were some cracks within the matrix, it seemed that the strength of the adhesion bond was strong enough to overcome the defect. The failed sample is shown in Figure 9 (c). According to the tensile test result, the sample started to decrease in tensile strength at the 40 wt% fibre loading. By observing Figure 9 (c), the PALF were not fully covered by the corn starch. Many gaps can be seen between the fibre and matrix.

5. Conclusions
As a conclusion, physical properties are affected by fibre loading, whereby the higher the fibre loading, the higher the value recorded for each test (density, water absorption, and moisture content). This means that for the density, water absorbtion and moisture content, the percentage of fibre loading
are affected directly by the fibre loading. This can be proven by comparing the results of 5 wt% fibre loading with the 70 wt% fibre loading, whereby the latter fibre loading had higher values for the three physical testings. The optimum flexural test was at fibre loading of 30 wt% for both flexural strength and modulus of elasticity as compared to the others. These findings were supported with the morphological analysis, whereby the internal particle of the PALF-reinforced corn starch was performed. The bond between the fibre and the matrix was observed through this analysis. Therefore, it could be concluded that for this research, the 30 wt% of fibre loading is the most optimum fibre loading for the 3cm length PALF as compared to the other composition ratios previously stated. The mechanical properties of the composites were also affected by the percentage of void and interfacial bonding between the PALF and corn starch.

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