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# Recycling of Pineapple (*Ananas comosus*) Leaf Agro-waste as One of the Raw Materials for Production of Eco-friendly New Paper

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**Abstract.** This project aimed to recycle pineapple (*Ananas comosus*) leaf agro-waste and wastepaper into new paper using simple processing routes that involve pulping, mixing, sieving, compaction and drying. Subsequently, physical and mechanical properties, as well as the morphology of the pineapple leaf fiber (PALF) -based paper were investigated accordingly. Two different samples were prepared whereby the first sample is pineapple leaf fiber mixed with wastepaper, and another sample is pure pineapple leaf fiber paper. The samples were tested for tensile properties by using the universal testing machine, morphological analysis using scanning electron microscopy, and density measurement using a densitometer. It was found that the sample consist of pineapple leaf and wastepaper shows a higher tensile strength than the sample containing only PALF. Moreover, the density of paper with pineapple leaf fiber and wastepaper mix was higher than the paper with only pineapple leaf fiber. In the future, further modification of body formulation is necessary to further improve the properties of the pineapple leaf fiber-based paper.

## 1. Introduction

Paper is generally regarded as a sheet constituting of cellulosic fibers, which are normally produced by separating wood cells by either mechanical or chemical means [1]. In the pulp and paper industry, hardwood and softwood are the main raw materials for sources of cellulose pulp. Wood contributes up to 90% of traditional raw materials for paper manufacturing around the world. However, forest tree depletion harmfully affects the environment as well as human health [2]. Therefore, it is crucial to control the utilization of conventional wood pulps for manufacturing of new paper. Additionally, the demand for paper keeps increasing over the years, making the current source insufficient [3].

Therefore, the industries opted out for alternative sources from a non-wood plant fiber, which are available in abundance as agro-waste material. Non-wood raw materials are used in the papermaking industry for less than 10% of total production for the industry worldwide [3]. Many benefits can be achieved as a result of utilizing pulp fiber, especially in terms of the economy and environment, since it can be found in abundance and hence is a sustainable resource [4]. Previously, pulp from empty fruit bunch, banana fiber, and carpet grass has been commercialized in the industry [5–7].



Pineapple (*Ananas Comosus*) leaf fiber, or also known as PALF is among the natural fibers that have the high cellulose content up to 81.27%, with minimal hemicellulose and lignin content at 12.31% and 3.46%, respectively. It has a similar density with other natural fibers. However, it has higher Young's modulus and ultimate tensile strength than did other natural fibers. Previous research has reported that PALF originated from different cultivar would contain higher cellulose content than did wood fiber [2]. PALF has high stiffness and specific mechanical strength. It has high cellulose compound and this lead to its superior hydrophilic characteristic. Fresh pineapple leaves could yield approximately 2 to 3% of PALF. The PALF consists of thin and small multicellular fibers and looks alike a thread. These cells are tightly joined with the aid of pectin compound. Considering the overall outstanding properties of PALF, industries can use them as an excellent alternative greener raw material to manufacture new paper instead of relying on the traditional pulps of wood. PALF is proven to have excellent mechanical strength, but due to lack of knowledge and experimental works, it is still not fully utilized and recycled into new paper production [1].

Waste paper is usually recycled and mixed with virgin fibers to produce new paper [8]. The pulp containing recycled paper has its advantages by using lignocellulosic fibers as additives are their low density, low cost, nonabrasive nature, high filling levels possible, low energy consumption, and a wide variety of fibers available throughout the world [9]. The use of pineapple leaf fiber (PALF) can be considered relatively new in the paper manufacturing industry in Malaysia. Hence, the purpose of this project is to observe the mechanical properties of the recycled paper utilizing PALF as an additive and to compare the characteristic between two types of paper which is made from only PALF and the mixture of PALF and waste paper which from here on referred to as PALF and PALFWP.

## 2. Methodology

### 2.1. Materials

Pineapple (*Ananas Comosus*) leaves were obtained from a local pineapple plantation farm located in Besut, Terengganu, Malaysia. Wastepaper from used papers, old papers, and foolscap papers was collected at the surrounding of Universiti Malaysia Kelantan Jeli Campus. Sodium bicarbonate ( $\text{NaHCO}_3$ ) powder was used for delignification [4].

### 2.2. Pineapple Leaf Pulping Process

Pineapple leaves were cut into small pieces and air-dried for 24 hr to remove moisture before boiled at 90°C for 90 min in 8L of distilled water with the addition of sodium bicarbonate powder. Sodium bicarbonate to pineapple leaves weight ratio was set as 1:4, according to previous research reported by Sibaly and Jeetah [10]. After boiling, 300 ml of boiled pineapple fiber slurry was added to 700 ml of water and liquidized for 5 min until the puree-like texture of pineapple leaves fiber (PALF) was formed.

### 2.3. Pineapple Leaf Fiber and Wastepaper Mixing Process

Wastepaper was cut into small pieces and liquidized for 5 min until puree texture was formed. After that, the smooth fiber was placed in a container and mixed with the prepared pineapple leaf pulp. The ratio for the pineapple leaf pulp to wastepaper was set as 1:1.

### 2.4. Sieving Process

Sieve was submerged into the prepared slurries of either PALF or PALFWP and shook to ensure even dispersion. The pulp was then carefully removed from the sieve. Further, the paper-shaped pulp was pressed under 3 tons compression using a hydraulic press to remove excess water. Eventually, it was dried for 24 hr at ambient temperature prior to testing.

## 2.5. Characterizations of PALF and PALFWP Paper

2.5.1. *Scanning Electron Microscopy (SEM)*. SEM micrographs were used to analyse the surface morphology and elemental distribution of the PALF and PALFWP papers by Scanning Electron Microscope, JSM-IT1000 (Japanese Electron Optics Laboratory Co. Ltd., Japan). SEM micrographs were acquired at various magnifications of 30x, 500x, 1000x and 2000x.

2.5.2. *Tensile Test*. Tensile test of new papers was performed using Universal Testing Machine following ASTM D 828-97 standard. Samples were prepared by cutting the paper 18 cm x 6 cm rectangular strips.

2.5.3. *Density Calculation*. The density of the paper produced was calculated using the following equation:

$$\rho = m/v \quad (1)$$

where,

$\rho$  = density,

$m$  = difference between mass of specimen before and after submerging in liquid, and

$v$  = volume of liquid

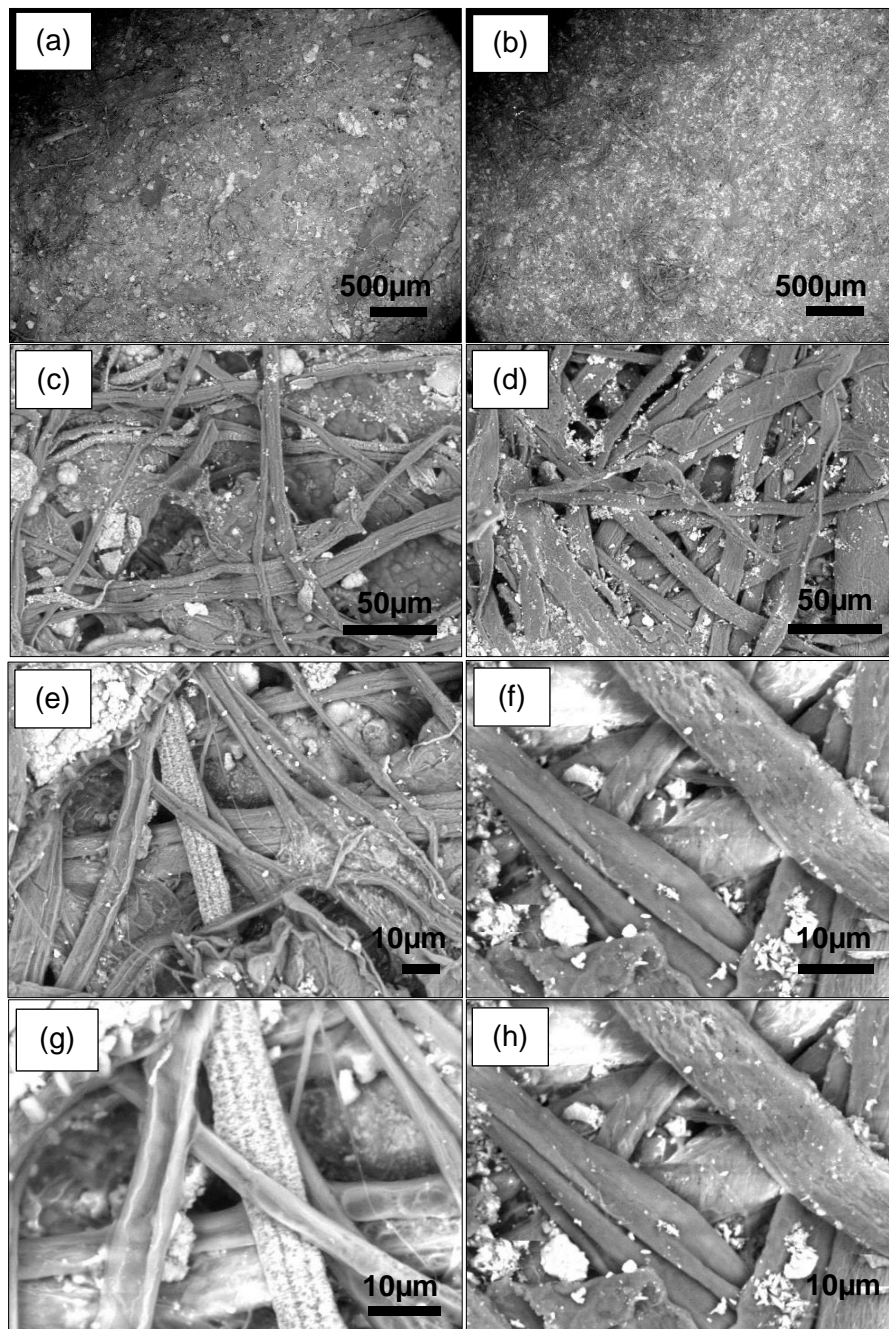
## 3. Results and Discussion

### 3.1. Morphology

Samples of PALF and PALFWP paper were subjected to SEM analysis. Figure 1 (a)-(h) illustrates the structure of the two samples at various magnifications. It can be observed that the paper made up of only PALF shows bundles of packed fiber, which was formed by elementary or ultimate cells overlapped and bonded together by lignin along the bundle length. The microstructure of PALFWP paper consisted of a closely packed fiber structure, and this could indicate that the strength of the PALFWP paper is comparable with PALF paper. Reddy and Yang (2005) have reported that the packing arrangement of fiber-matrix on the surface of pineapple leaf increase mechanical strength of new paper [11]. This fully aligned with the microstructural findings of this study.

In addition, the microstructural analysis also shows that the cellular structure of fibrils was connected with plant tissues and to form fibers of the papers. Similar to most lignocellulosic fibers, PALF showed irregular cross-section and diameter variations of fibers. However, traces of waxy materials and certain defects occurred of the fiber surface due to over-pressure during the compaction process. The defects could further deteriorate the mechanical properties of the new paper.

The surface of fiber is containing waxes and other protective compound like lignin, pectin, and hemicelluloses. These compounds would then form a thick condensed and protective layer to protect the cellulose inside the fiber's matrix layer. This condensed and agglomerated structure is an important feature of the new paper derived from PALF [11]. From the observations as in Figure 1, both surfaces of the new papers are consisting of condensed arrangement of the fiber, as this is well align with the findings reported by Daud et al. (2014) [11]. As a result, the features would increase the strength of PALF and subsequently improving overall properties of the new paper produced. Furthermore, SEM images of the PALFWP sample show that fibers were surrounded by the matrix of wastepaper and the remaining fly ash particles. This ash content is a function of the absence or presence of other materials (various chemicals, metallic and mineral matters). The ash content in this sample is more than the pure pineapple leaves sample. Lower ash content would lead to good properties of new paper derived from PALF [11].

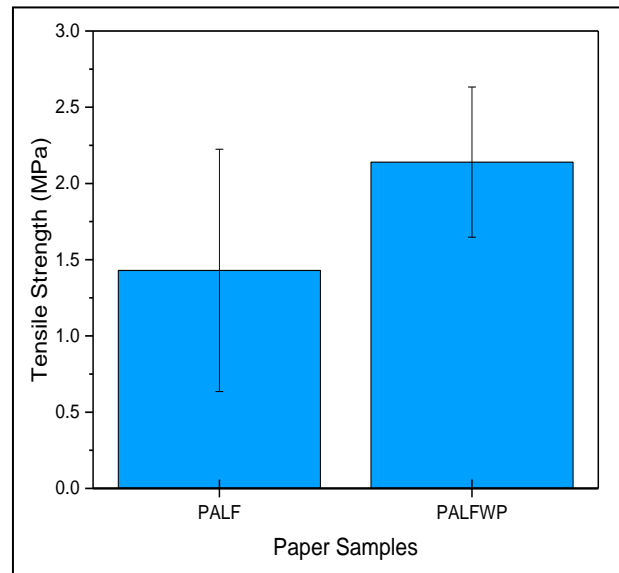


**Figure 1.** SEM Micrographs of Samples at Different Magnifications: (a) PALF at 30x (b) PALFWP at 30x (c) PALF at 500x (d) PALFWP at 500x (e) PALF at 1000x (f)PALFWP at 1000x (g)PALF at 2000x and (h) PALFWP at 2000x

### 3.2. Tensile Strength

Three samples were used for the tensile test of the recycled paper. Figure 2 shows that the average tensile strength of PALF sample is lower than PALFWP with 1.431 MPa and 2.140 MPa, respectively. The error bar shows some deviation that may occur due to the difference in thickness of the samples. It was found that the tensile strength of PALFWP paper was higher than the one with pure PALF paper due to the formulation containing waste paper consist of cellulose fiber, which will also act as reinforcing fiber in the new paper. It was also owing to the excellent fiber-fiber interaction between the PALF and waste

paper [10]. The superiority of pineapple leaves fibers mechanical properties are related to the high content of alpha-cellulose content and low microfibrillar angle ( $14^\circ$ ) and supported by additional compositions from used papers that make the recycled paper much more durable.



**Figure 2.** Tensile Strength of PALF and PALFWP Paper Samples

### 3.3. Density

The density of PALF paper was found to be lower than PALFWP paper with the values of 1.15 and 1.36  $\text{g/cm}^3$ , respectively. This can be related to the SEM micrographs that show the structure of the mix are more closely packed together with smaller gaps in between the fibers. This ensures a higher density to be achieved onto the recycled paper. Moreover, the tensile test also supported that the mixed sample has a higher strength. This may also be due to its high density, which in turn contributes to the more efficient strength improvement to the paper produced.

## 4. Conclusion

The morphological and tensile properties of the PALF and PALFWP paper shows an average of 1.431 MPa and 2.140 MPa, respectively. The results indicate that the combination of PALF with waste paper brought significantly higher tensile strength than PALF-only paper. This finding also supported by the discussion in Scanning Electron Microscope (SEM) and discussion in density to shows that pineapple leave fiber has higher tensile strength than paper with a mixture of waste paper. Morphology of PALF-mix paper shows that it is filled with many fiber matrices that are packed together and making the paper stronger. PALF paper, however, exhibits unique characteristics which are the presence of waxy surface and other elements such as lignin, pectin, and hemicelluloses. This characteristic gives the paper a similar behavior as the oil paper where the water absorption rate is low. This is a good substitute for the conventional oil paper that use petroleum product (plastic) that can pollute the environment.

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