



Pineapple leaf fiber reinforced wheat gluten biocomposites

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Abstract

In this work, pineapple leaf (PAL) fiber reinforced biocomposite materials were manufactured using a hot-press molding. Two conditions of PAL (fresh and dried leaves) were used as reinforcement; meanwhile, wheat gluten (WG) was used as a matrix phase. Bulk density of the biocomposite samples was measured, and then the flexural strength, strain at break and modulus of elasticity of the biocomposites as a function of PAL condition and particle size were investigated. Fiber-matrix adhesion, fiber breakage, and failure topology of the biocomposites were examined using scanning electron microscopy (SEM). It was found that the different conditions and particle sizes of PAL significantly affect mechanical properties of the resulting biocomposites. The dried PAL was shown to be superior to the fresh PAL as reinforcement in the composites due to a better interfacial adhesion with the WG matrix as confirmed by the series of SEM images. The biocomposites with large particle size of dried PAL showed the best mechanical performance which considered to be adequate for applications such as disposal food container and packaging.

Keywords: biocomposites, pineapple leaf fiber, wheat gluten, scanning electron microscopy, mechanical properties

Introduction

Due to the environmental concerns, many efforts are being made to develop composite materials from renewable resources. Composites reinforced with natural fibers such as sisal, banana, pineapple, jute, bamboo fiber and kenaf have been extensively studied and found to provide good reinforcement in polymer matrices (Li et al. 2000; Rana et al. 2003; Elbadry et al. 2012). Among these natural fibers, pineapple leaf (PAL) fiber shows excellent mechanical properties (Devi et al. 2011). The main chemicals consistent of PAL fiber are cellulose (70–82%), lignin (5–12%), and ash (1.1%). Mukherjee and Satyanarayana (1986) reported that PAL fiber possessed tensile strength and Young's modulus in range of 413–1627 MPa and 34.5–82.5 GPa, respectively. Besides the high mechanical performance, it has low density and also low cost. In Thailand, pineapple is considered as the top economic crops and produced higher than 1.8 million tons per year (Food and Agriculture Organization of the United Nations: Division of Statistics, 2014). It has been reported that PAL fiber was an effective reinforcement in polyester matrix (Devi et al. 1997). In addition, the systems of PAL fiber and biodegradable polymer such as poly(lactic acid) (PLA) have also been studied (Huda et al. 2006; Huda et al. 2008). However, there have been limited studied on properties of PAL fiber and biodegradable polymers derived from other resources. Wheat gluten (WG)

is known as a by-product of the starch industry which contains proteins more than 75%. It received increasing interest from many researchers, due to its attractive functional properties and biodegradability (Guillard et al. 2013). When mixing WG with plasticizers such as glycerol, a three-dimensional network material with good viscoelasticity can be created. (Payne and Corfield 1979). Moreover, WG-based materials can fully degrade without releasing toxic product, thus, are truly environmental friendly (Domenek et al. 2004). The present investigation was aimed at analyzing the structure and mechanical properties of PAL reinforced WG-based biocomposites. The influences of PAL condition (fresh and dried leaves), and particle size on the bulk density, flexural properties, and fracture surface of the biocomposites were analyzed.

Methodology

Materials

Fresh PAL was purchased from Nang Lae district, Chiang Rai, Thailand. Wheat gluten powder was supplied by Zhangjiagang Hengfeng Starch Products Co. Ltd., China. Glycerol was supplied by Unionscience Co. Ltd., Chiang Mai, Thailand.

Composite Processing

Fresh PAL (as received) was chopped using a blender to reduce size and sieved into two ranges of desired particle size (500-2500 μm and $>2500 \mu\text{m}$). After air drying the fresh PAL for 1 month, the dried PAL was prepared in the same way as did for the fresh PAL. All biocomposites were prepared by, firstly, mixing wheat gluten powder, glycerol, and PAL particles together by hand in a bowl at room temperature for roughly 15 min. The ratio of WG/glycerol/PAL in the biocomposites was fixed at 3.5/1.5/5 by weight. The mixture was then placed between two transparent polyethylene terephthalate (PET) sheets and compression molded (Labtech LP-S-80) at 130°C and 60 tons for 10 min. After compression, the resulting composite was cooled down for 5 min and removed.

Characterization

Bulk density

For raw materials, PAL particles were free-flowing filled into the volume container of 416.7 cm^3 (V) and then the mass of PAL particles, M (g) was measured using a 2-digit electrical balance. The bulk density (D) in g/cm^3 was calculated using the following equation:

$$D = \frac{M}{V} \quad (1)$$

For biocomposites, the samples were cut into a dimension of 8.2 cm \times 2.0 cm. Then, the samples were stored in the controlled relative humidity container of 59.2 \pm 0.2% (using saturated $\text{Mg}(\text{NO}_3)_2$ solution) for not less than 40 hours at 25°C. After that, the mass of samples, M (g) was measured using a 2-digit electrical balance. The length, width and thickness of the samples were then measured using a digital vernier caliper to determine the

sample volume, V (cm^3). The bulk density of biocomposites, D (g/cm^3) was estimated using the equation (1).

Mechanical property measurement

The biocomposites were cut into a specific dimension according to ASTM D6109-13. Then, the samples were conditioned at $59.2\pm 0.2\%$ relative humidity for not less than 40 hours at 25°C before testing.

Flexural tests (3-point bending mode) of the composites were performed using universal testing machine (UTM, INSTRON®, 5560 series dual column system, USA) equipped with a 1 kN load cell. The cross head speeds were fixed to 2 mm/min. The sample is deflected until rupture occurs or until the maximum fiber strain of 3% is reached, whichever occurs first. Modulus of elasticity, flexural strength and elongation at break were evaluated from at least five duplicates for each sample.

Surface morphology

The PAL particles and fracture surface of biocomposites were directly observed after gold coating. Scanning electron microscope (SEM, LEO model 1450 VP, ZEISS, UK) was operated at 10 kV.

Results and Discussion

SEM images of different conditions and particle sizes of PAL are shown in Figure 1. Both fresh and dried PAL with particle size of $500\text{-}2500\ \mu\text{m}$ were mostly flakes, possibly broken away from the surface of PAL during chopping (Figure 1a and b, respectively). On the other hand, for the PAL particles large than $2500\ \mu\text{m}$, both fresh and dried PAL particles were more like a group of long fibers partially hold together (Figure 1c and d). The fibers in the dried PAL of large particle size ($> 2500\ \mu\text{m}$), however, seemed to be more exposed and separated into individual fibers (Figure 1d).

The bulk density of PAL particles (a raw material) and biocomposites were listed in Table 1. The bulk density of PAL with small particle size ($500\text{-}2500\ \mu\text{m}$) is greater than that of the large particle size ($>2500\ \mu\text{m}$). This should be because small particles, in general, could be moved and packed easily. In case of biocomposites, bulk density of all samples was found to be in the same range. Thus, condition and size of PAL particles does not affect the bulk density of the biocomposites.

Table 1: Bulk density of PAL particles and biocomposites

Type of particle	Bulk density of raw material (g/cm^3)	Bulk density of biocomposites (g/cm^3)
$500\text{-}2500\ \mu\text{m}$, fresh leaf	0.151 ± 0.008	1.339 ± 0.013
$500\text{-}2500\ \mu\text{m}$, dried leaf	0.142 ± 0.005	1.301 ± 0.027
$>2500\ \mu\text{m}$, fresh leaf	0.074 ± 0.003	1.302 ± 0.068
$>2500\ \mu\text{m}$, dried leaf	0.071 ± 0.004	1.308 ± 0.017

The mechanical properties of biocomposites are given in Table 2. For biocomposites reinforced with both sizes of the fresh PLA, the flexural strength lies between 5.95 and 6.53 MPa. While the biocomposite reinforced with the large particles of dried PLA shows the highest strength (11.56 MPa) when compared to the others. For PAL of the same particle sizes, it is obvious that the biocomposites reinforced with the dried PAL exhibited the higher flexural strength. In addition, for the biocomposite with the dried PAL, the sample reinforced with the small particles showed the higher modulus than the one with large particles. Strain at break can be observed only in this composite sample because the others did not fracture at 3% strain.

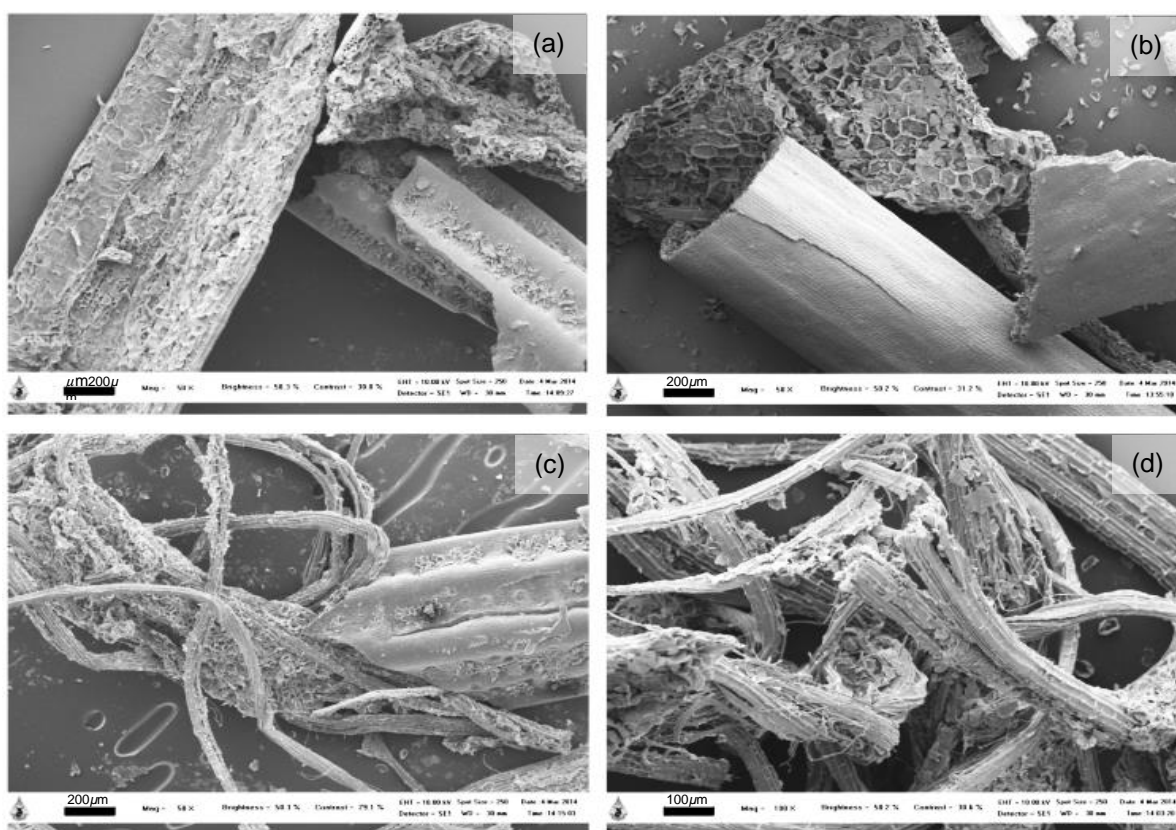


Figure 1: (a) Fresh and (b) dried PAL with particle size of 500-2500 μm ; (c) fresh and (d) dried PAL with particle size greater than 2500 μm observed by scanning electron microscopy.

Table 2: Mechanical properties of biocomposites reinforced with PAL

Type of particle	Strength (MPa)	Modulus (MPa)	Strain at break (%)
500-2500 μm , fresh leaf	6.53 \pm 0.28	536.01 \pm 28.98	*
500-2500 μm , dried leaf	9.70 \pm 0.52	805.47 \pm 59.80	2.44 \pm 0.27
>2500 μm , fresh leaf	5.95 \pm 0.22	461.28 \pm 22.13	*
>2500 μm , dried leaf	11.56 \pm 0.79	766.47 \pm 62.92	*

*The specimens did not fracture at 3% strain.

Figure 2 shows the tensile stress-strain curves of all biocomposites. It can be observed that all samples exhibit linear region at low strain and a change in slope of the curve is observed at high load. From these curves, it is further confirmed that the dried PAL particles were superior to the fresh PAL particles as reinforcement in the WG-based composite materials. During the composite preparation, it was observed that after drying the chopped PAL particles, the fresh PAL condition seemed to be clumped together and formed the larger aggregates when compared to the dried PAL particles for both sizes. This large PAL aggregates possibly leads to a poor distribution of PAL as well as a poor interfacial adhesion within the composites. Thus, these might be the reasons for low mechanical properties of the composites reinforced with the fresh PAL particles versus to those reinforced with the dried PAL particles.

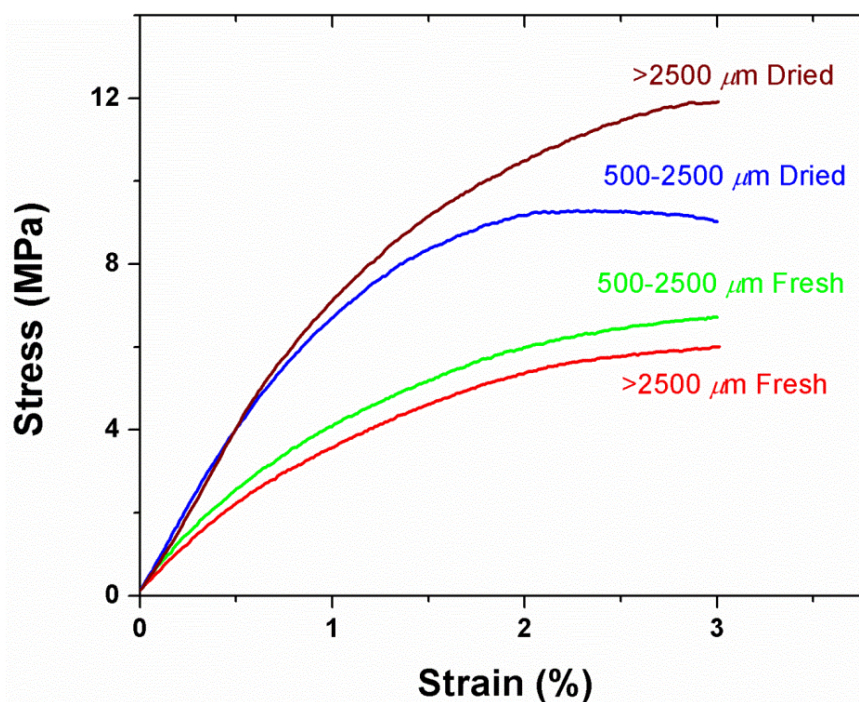


Figure 2: The relationship of stress-strain curves of biocomposites from PAL.

The fractured surfaces of the biocomposites observed by scanning electron microscopy are shown in Figure 3. For the biocomposites reinforced with small PAL particles (both fresh and dried types), it was observed that the PAL flakes were distributed as layers and surrounded by the WG-based matrix. Fiber pull-out and fiber split were barely observed in these samples (Figure 3 a-d). In case of the biocomposites reinforced with the large size of both fresh and dried PAL particles, fiber pull-out, debonding and fibrillation were clearly observed on their fracture surfaces (Figure 3 e-h). However, in the biocomposites reinforced with the fresh PAL particles of large size, fiber were extensively and cleanly pull-out. No matrix phase was found to bonded or adhered on the pull-out PAL particle surfaces. This indicated to a poor interfacial adhesion between the fresh PLA particles and the WG matrix phase which consequently led to inferior mechanical properties of this composite.

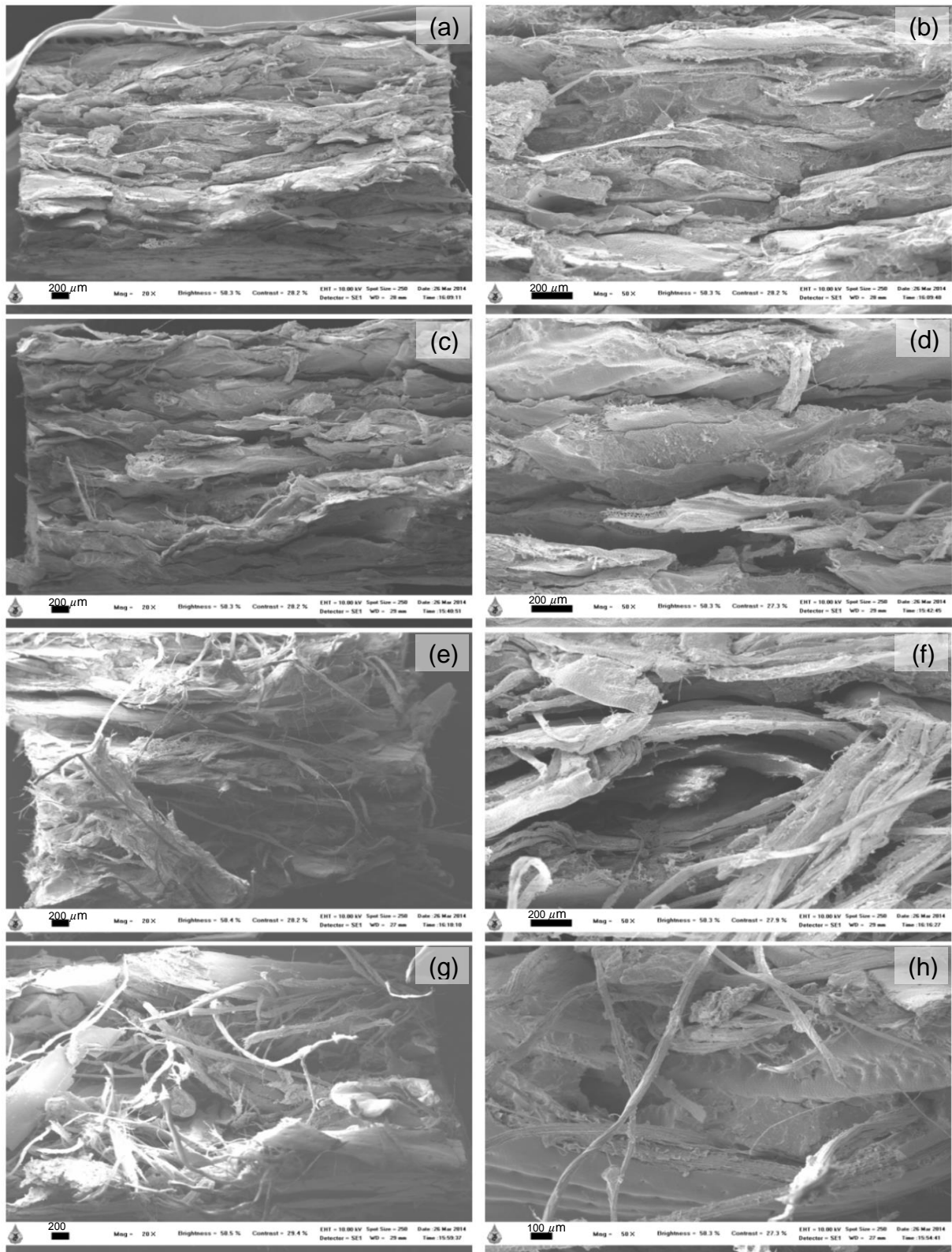


Figure 3: Fracture surface of biocomposites from fresh (a-b) and dried PAL (c-d) of particle size 500-2500 μm ; fresh (e-f) and dried PAL (g-h) of particle size greater than 2500 μm observed by scanning electron microscopy.

Conclusion

Both fresh and dried PAL with particle size of 500-2500 μm were flakes. On the other hand, for the PAL particles larger than 2500 μm were more like a group of exposed long fibers. In this work, the WG-based biocomposites reinforced with fresh and dried PAL particles (500-2500 μm and $>2500 \mu\text{m}$) using glycerol as a plasticizer can be successfully prepared by hand mixing and then thermo-molding at 130°C. It was found that the bulk density of WG-based composites reinforced with all PAL types was in the same range around 1.30-1.34 g/cm^3 . This indicated that condition and size of PAL particles were not affect the bulk density of the biocomposites. For mechanical properties, the system reinforced with the large particles of dried PLA showed the best performance when compared to the others. For the same size of PAL particles, it is proved that the dried PAL particles were superior as reinforcement in the WG-based composite materials. SEM revealed that, in case of the biocomposites reinforced with fresh PAL particles of large size, clean fiber pull-out was clearly observed, indicating to a poor interfacial adhesion with the WG matrix which consequently led to a reduction in mechanical properties of this composite. The mechanical properties of this biocomposites can be further improved by various surface modification methods.

Acknowledgements

This work was granted by the National Research Council of Thailand (NRCT) and Mae Fah Luang University, Chiang Rai, Thailand. The authors wish to thank STIC staff for their assistances in mechanical tests and SEM.

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