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Self-Cleaning and Hydrophobic Pineapple Peel Fibre based Biocomposite

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Abstract. Biocomposite has been widely used as plastics replacement for its biodegradability. However, the water absorptivity defeats its purpose as food packaging. The water absorption analysis for pineapple peel fiber (PAPF) was done according to ASTM D570-98 and it has been proven that the water content increases to 7.2% with the 50% content of PAPF in the bio-composite (50% PAPF). For that matter, hydrophobic coating was synthesized and applied on PAPF based bio-composite. The bio-composite was produced using Low Density Polyethylene (LDPE), pineapple peel fibre (PAPF), Linear LDPE grafted maleic anhydride (LLDPE-g-MA) as compatibilizer and Refined, Bleached Deodorized Olein (RBDOL) as plasticizer. The coating was synthesized by using polydimethylsiloxane (PDMS) and decamethylcyclopentasiloxane as hydrophobic component, silica nano-particles as nanostructured particles and non-organic solvent and non-ionic surfactant p-octyl polyethylene glycol phenyl ether as surfactant. The emulsion was homogenized and after that sprayed onto the surface of PAPF bio-composite and dried in the oven at 50-60°C for 24 hours. Through the 100 μ m magnification using field emission scanning electron microscope (FESEM), the hydrophobic coating was proven to conform the Cassie-Baxter hydrophobic property. C-O bond from the PAPF disintegrates after the production of bio-composite from the Attenuated Total Reflectance Fourier Transform Infrared (ATR FTIR) spectrometry transmittance. The application of the coating onto the surface of PAPF bio-composite gave the surface hydrophobic property where the contact angle changes from 82.27 \pm 2.66° which is hydrophilic to 122.63 \pm 2.17° which is hydrophobic. The bio-composite with the highest PAPF content will also exhibit self-cleaning ability which makes it suitable for the usage of hydrophobic food packaging material.

1. Introduction

The extensive production of conventional plastics and their use in different commercial applications poses a significant threat to both the fossil fuels sources and the environment. Alternatives called bio-composite evolved during development of renewable resources. Natural fibre-based bioplastic from



agricultural crop waste is interesting alternative as it is cost effective and renewable resources type of bioplastic. However, hydrophilic nature of natural fibre-based biocomposites will lead to high moisture content of bioplastic, thus reducing its physical and thermal properties and consequently limit its application [1]. Therefore, 'lotus effect' characteristics is introduced to overcome the weakness of natural fibre-based plastics towards water content and moisture. This research was proposed to formulate high content of pineapple peel fibre (PAPF) bio-composite with the addition of 'lotus-effect' special characteristics to enhance its application. 'Lotus-effect' is known as the self-cleaning ability due to the hydrophobic properties [2]. This means that the surfaces can repel contaminants such as solid particles, organic liquids, and biological contaminants with a hint of water drop [3]. The modification of hydrophobic solution was prepared by mixing polydimethylsiloxane, silica nano-particles and non-ionic surfactant and applied to PAPF bio-composite. Characterization and self-cleaning performance analysis were studied using infrared spectroscopy, water contact angle, scanning electron microscopy and water resistance analysis. Self-cleaning surfaces fibre-based bioplastic with a very high static water contact angle and hydrophobic characteristic have potential to be commercialized in the field of utensils, exterior/interior parts of bulk water by almost 30%. With the hydrophobic property, water droplets with the contact angle of about 120°-150° will be able to roll-off the super hydrophobic surface at 10° inclination. Particulates on top of hydrophobic surfaces will roll-off along with the water, giving the surface a 'self-cleaning' ability. This 'self-cleaning' ability works due to the adhesion between water droplet and particulate is stronger than the water adhesion to the surface. Application of this self-cleaning ability for outdoor kitchenware or electronic devices can be very useful.

2. Experimental

PAPF (dried pineapple peel from Lee Pineapple Co. (Pte) Ltd, *Malaysia*) and LDPE (TITANLENE® LDF 260GG) were dried at 60°C under desiccant drier to prevent from void moisture and to ensure moisture removal for 24 hours prior to compounding with additional cooking oil as plasticizer and LLDPE-g-MA (*DuPont Malaysia*) as compatibilizer. PAPF was compounded with LDPE by using co-rotating twin extruder (SINO PSM 30) with the feed temperature at 110°C, then compressed at 120°C, metered at 120°C and die zoned at 110°C at the speed of 80 rpm. The feed section of the barrel was fed with pre-mix of PAPF (with different percentage of fiber ranging from 10%, 20%, 30%, 40% and 50% wt) with LDPE. The shearing action of the machine melted and homogenized the PAPF and LDPE. The strands of PAPF bio-composites were then passed through palletizer machine to produce PAPF bio-composites resin. Silica nanoparticles were first dispersed into PDMS and then followed by the addition of polydimethylsiloxane (silicone oil) into the mixture. Surfactant was added into the mixture dropwise with a dropper and stirred further for 10 minutes using magnetic stirrer with mass ratio of 3:57:39:1 respectively. Water absorption test was carried out to determine the water absorptivity of samples with different PAPF content in biocomposite. Samples were then characterized using Fourier Transformed Infrared spectroscopy (FTIR) (*Perkin Elmer*), Field emission scanning electron microscopy (FESEM) (JSM-7800F, *JEOL*), water surface angle analysis (*VCA Optima*) and soil burial test.

3. Result and discussion

3.1. Water Absorption Test

Water absorption test was carried out to determine the water absorptivity of samples for different concentration before and after the hydrophobic coating. Figure 1 illustrates the water content for 10% PAPF, 20% PAPF, 30% PAPF, 40% PAPF and 50% PAPF. For LDPE, the water content did not increase due to its water resistivity. Figure 1 shows the higher the percentage of PAPF in the bio-composite, the higher the water content. PAPF contains carboxymethyl cellulose which has good water absorption [4]. 50% PAPF biocomposite showed water absorption up to 7.2% after 30 days immersed in water; which is not suitable for food packaging. The water absorption issue has been solved after the application of hydrophobic coating. The hydrophobicity changes the PAPF bio-

composite morphology as proven by the FESEM result which makes it harder for water to adhere to the surface due to the conformance to the Cassie-Baxter state of wetting.

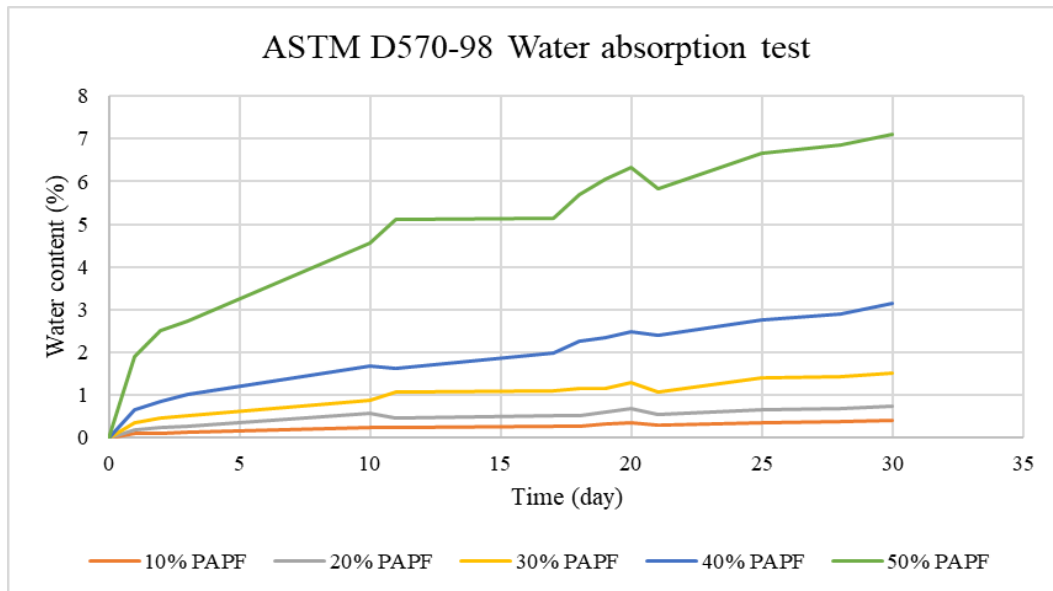


Figure 1. Water absorption test

3.2. Fourier Transform Infrared (FTIR) Spectroscopy

Fourier Transform Infrared (FTIR) Spectroscopy was conducted on LDPE, PAFP and PAFP biocomposite with 50% wt of fiber content (50% PAFP) to identify the functional group that exists on the sample using ATR mode. Figure 2 illustrates the transmittance spectrum of LDPE, PAFP, and 50% PAFP. From Figure 2 it can be concluded that the functional group in LDPE is only C-H group due to the existence of the peak at 1465 cm^{-1} which represents C-H (methyl) group. The spectral band of LDPE shows the same result as Moreno et al. (2018) [5]. For pineapple peel fibre, the spectral shows peak at 1414 cm^{-1} and 2931 cm^{-1} which represents C-H (methyl) group, 3303 cm^{-1} for O-H (alcohol/phenol) group, 1038.71 cm^{-1} for C-O (alcohol) group and 1630 cm^{-1} for C-C bond. The same spectral result was obtained by Yob (2017) [5] for PAFP. Dai et al. (2017) [4] stated that pineapple peel fibre contains carboxymethylcellulose, which is, $\text{C}_8\text{H}_{16}\text{O}_8$. Whereas for 50% PAFP, the peak existence at $1464\text{--}1156\text{ cm}^{-1}$, 2850 , 2917 cm^{-1} represents C-H (methyl) group, 3336 cm^{-1} represents O-H (alcohol/phenol) group, 1645 cm^{-1} represents C-C bond and finally 1030 cm^{-1} represents Si-O group. Although this slightly differs from the result obtained by Yob (2017) [6]. The absence of carbonyl groups in LDPE and PAFP/LDPE bio-composite denotes that the degradation after thermochemical processing in producing the bio-composite. According to Moreno et al. (2018) [5], this condition has proven that the carbonyl group absence indicated the polymers degradation. Dispersion of nanoparticles in a base fluid not only contributes to enhancement of thermal conductivity, but also because of greater heat transfer area, superior convective heat transfer coefficient can be achieved, which will also lead to enhancement of heat transfer.

3.3. Field emission scanning electron microscopy

The morphology of pineapple peel fibre before and after hydrophobic coating were observed by using Field Emission Scanning Electron Microscope (FESEM). Due to the nonconductive properties of the samples as it does not contain metal, a layer of platinum was coated on the samples to avoid the sample from quickly being charged. Figure 3 shows the micrographs of 50% PAFP bio-composite before and after the application of hydrophobic coating. The surface of bio-composite is more uneven due to its velvet-like surface; for sample with hydrophobic coating. This pile weave microstructure

gives the surface of the sample an uneven feel mimicking the Lotus leaf structure [3]. With that, any water droplet on top of the surface is proven to conform the Cassie-Baxter state of wetting which is verified by the water surface angle analysis.

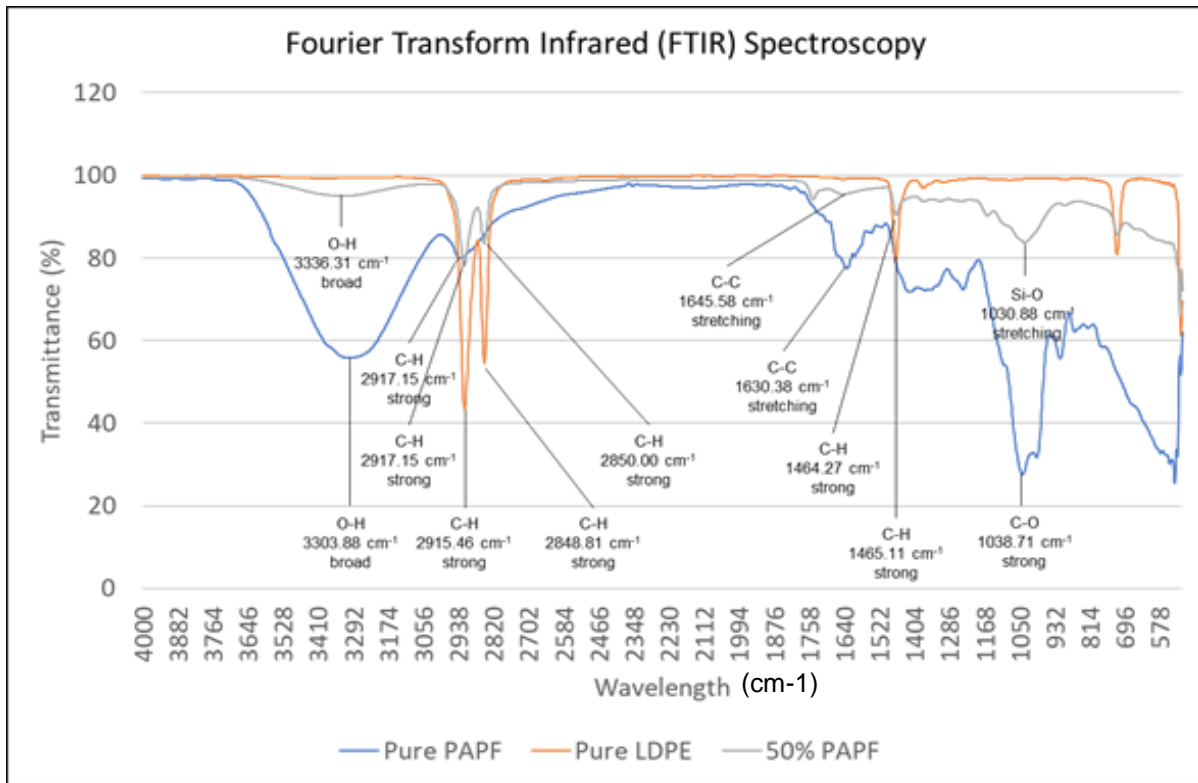


Figure 2. Fourier Transform Infrared Spectroscopy analysis

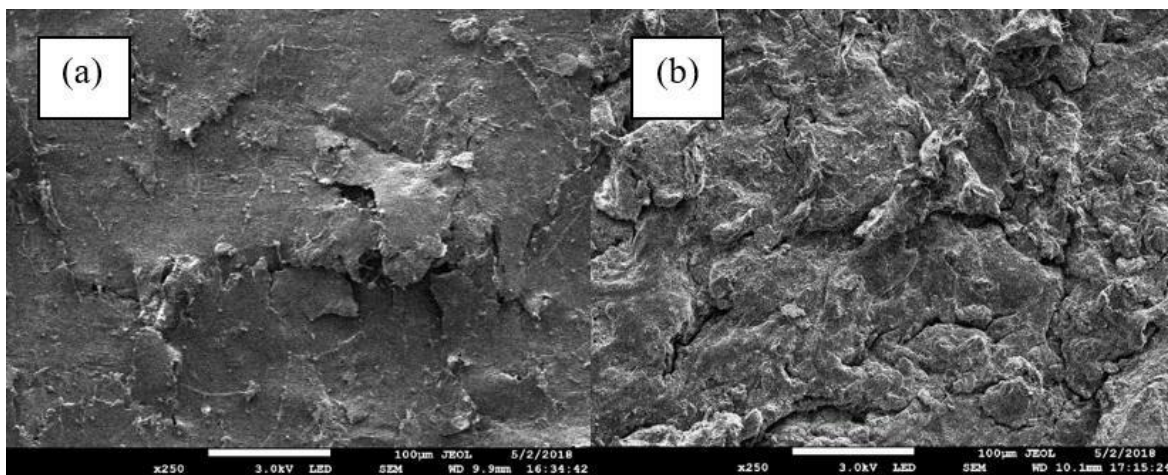


Figure 3. FESEM micrographs of 50% PAPF (a) before (b) after hydrophobic coating

3.4. Water Surface Analysis

Water contact angle test was conducted on the 50% PAPF sample before and after the application of hydrophobic coating. Figure 3 shows the images with the contact angle indication at the top left of the

image. Table 1 summarises the contact angle result for both before and after the hydrophobic coating. From Table 1, the application of the coating increased the water contact angle from $82.27 \pm 2.66^\circ$ which is hydrophilic to $122.63 \pm 2.17^\circ$ which is hydrophobic. The changes occur due to the change in the morphology of the 50% PAPF as can be seen in Figure 4. The result could be further improved if the morphology after the hydrophobic coating mimics the surface of the lotus leaf. This finding was supported by FESEM and Cassie-Baxter result, whereby PAPF bio-composite with hydrophobic coating showed higher contact angle. This Lotus-like bio-composite is water resistant and suitable for the usage of food packaging.

Table 1. Water surface contact angle analysis

Test	Water Contact Angle			
	Before Coating		After Coating	
	Left	Right	Left	Right
Test 1	85.00	87.40	121.20	121.90
Test 2	80.10	80.10	126.30	125.40
Test 3	80.70	80.30	120.80	120.20
Average	81.93	82.60	122.77	122.50
Total	$82.27 \pm 2.66^\circ$ (hydrophilic)		$122.63 \pm 2.17^\circ$ (hydrophobic)	

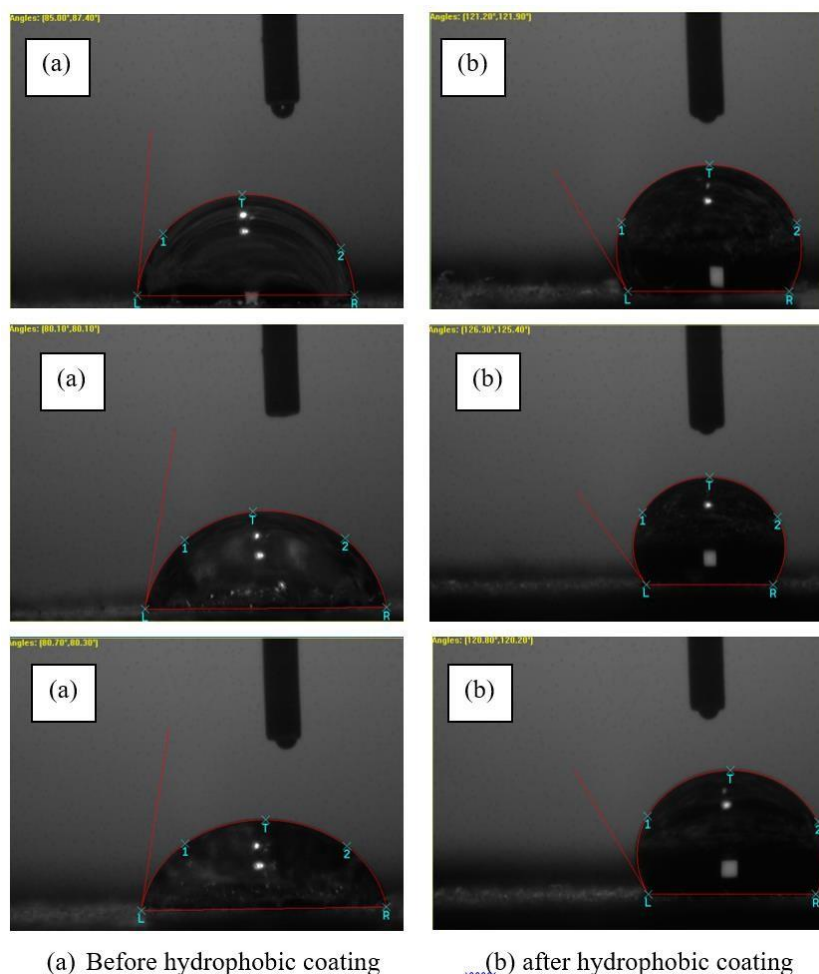


Figure 4. Water surface contact angle analysis

4. Conclusion

In conclusion, the application of the coating onto the surface of 50% PAPF bio-composite gave the surface hydrophobic property, whereby the contact angle changes from $82.27 \pm 2.66^\circ$ which is hydrophilic to $122.63 \pm 2.17^\circ$ which is hydrophobic. It was also proven to portrait the Cassie-Baxter hydrophobic pattern from FESEM analysis. The biodegradability study was carried out using soil burial analysis, and it showed that higher content of PAPF biocomposite showed higher percentage of weight loss content. Thus, the 50% PAPF biocomposite with the will exhibit self-cleaning ability which makes it suitable for the usage of hydrophobic food packaging material.

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