Physicochemical properties of *Artocarpus heterophyllus* peels packaging paper coated with fish gelatin-palm wax-lemongrass oil

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Abstract

Paper demand in the Philippines is increasing by 2.5% each year since 2016, including packaging paper. To address this demand, paper made from *Artocarpus heterophyllus* peels (AHP) has been used for packaging but it had low water barrier properties. A coating composed of fish gelatin, palm wax, and lemongrass essential oil (GPL) increases the physical, mechanical, and functional properties of paper. This study aims to determine the physicochemical properties of packaging paper made out of AHP coated with GPL. This includes water absorption, tensile strength, and chemical composition. Results showed that the water resistance of AHP-GPL was weaker than other packaging papers. The tear index was lower than kraft but possessed similar values with other non-wood papers. Attenuated total reflectance - Fourier Transform Infrared Spectroscopy (FTIR) results revealed that the peaks closely correspond to cellulose, which is a primary component of paper. This study concludes that AHP-GPL does not comply with paper packaging standards in water absorption and tear index.

Introduction. - The total annual demand for paper in the Philippines has increased by 2.5% every year since 2016, with the packaging paper industry experiencing a high growth rate [1]. Packaging paper possesses low moisture resistance due to its composition, allowing moisture to pass through and contaminate the contents [2] easily. To combat this weakness, coatings are added to increase their water barrier and mechanical properties [2].

Artocarpus heterophyllus (AH), commonly known as jackfruit, holds significant importance as a valuable crop in the Philippines [3, 4]. AH peels are often discarded as solid waste. According to the Philippine Statistics Authority [5], an average of 2,406 metric tons of AH waste was produced from 2019 to 2021 per year. The biodegradation of AH peels through anaerobic digestion produces air pollutants such as methane and carbon dioxide [6].

Lothfy et al. [9] tackles the use of *Artocarpus integer* peels as paper, and are compared to the standards of commercial paper. Results present that it has good mechanical properties but possesses low wet tensile strength, strain, and tensile energy absorption characteristics comparable to wood-based paper. It does not tackle other parameters such as biodegradability and water absorption. The use of *Artocarpus heterophyllus* paper (AHP) in packaging could be increased by enhancing its current features by addressing the weakness revealed in prior studies and boosting water resistance and other characteristics. Syahida et al. [7] conducted a study on the effects of kraft paper coated with an FG-PW-LEO (GPL) coating. The coating was made out of fish gelatin (FG), palm wax (PW), and lemongrass essential oil (LEO). FG has great potential for application as a paper coating due to its biodegradability and good film-forming properties [8]. PW is preferable due to its low cost, high availability, nontoxicity, excellent hydrophobicity, and biodegradability [7]. The lemongrass essential oil will act as the antioxidant property of the coating. The results prove that GPL coating can improve the water barrier, biodegradability, and mechanical properties of paper.

The validity of materials is often confirmed through Attenuated total reflectance - Fourier transform infrared spectroscopy (ATR-FTIR). Studies, such as that of Garside & Wyeth's [10], use ATR-FTIR spectroscopy to characterize and identify functional groups found not only in paper [11,12] but also in its components such as cellulose and lignin [13,14].

Based on the standards of the Technical Association of the Pulp and Paper Industry (TAPPI) 441, water absorption is evaluated by measuring the weight of water absorbed in a period of time using a Cobb tester. Syahida et al. [7] tested the water absorption of kraft paper with GPL coating. The coated kraft paper had lower water absorption values which indicates better water resistance [7].

Meanwhile, tear resistance evaluates the strength of a paper [15]. The study of Lothfy et al. [9] measured the tear index of *Artocarpus integer* paper.

The value of the tear index they obtained was lower than the commercial paper they compared. The tear index can be obtained by dividing the tearing resistance by the grammage of the paper [15].

Exploring the capabilities of AHP with different coatings and studying more parameters can help determine the most effective coating that can amplify the properties of the AHP. The study's outcome can provide additional information about more ways to reuse AH wastes rather than letting AH cause pollution. In addition, this can determine if AHP-GPL can help address the rise of paper demand. It can also encourage and further the development of AHP as an alternative packaging material. Moreover, this can help the environment by turning *Artocarpus heterophyllus* peels into paper rather than letting them biodegrade and cause pollution

This study aims to determine the physical and mechanical properties of packaging paper made out of *Artocarpus heterophyllus* peels (AHP) coated with Fish Gelatin-Palm Wax-Lemongrass Oil (GPL) and to be compared with the respective kraft paper standards. Testing the AHP coated with GPL coating (AHP-GPL) will help evaluate if the GPL coating can address the low barrier water properties of the AHP paper and determine if it can be on-par or even better than the given standard for papers.

Specifically, the aims of this study are to:

(i) identify functional groups found within the AHP produced via Attenuated Total Reflectance Fourier-Transform Infrared (ATR FTIR) spectroscopy, and ensure the validity via comparison to literature;

(ii) measure the water absorption (WA) of GPL-coated AHP (AHP-GPL) via the Cobb Test, and compare to corresponding standards; and

(iii) determine the tear resistance (TR) of AHP-GPL by using a tear tester, and calculate the TI for comparison to existing literature.

Methods. - Papermaking followed a modified soda pulping process to extract pulp from *Artocarpus heterophyllus* peels [9]. The mold and deckle used are 27 cm by 22.5 cm in length and width, allowing each sheet of paper to form one sample for the Cobb test. The preparation of the coating was made in the same process as in Syahida et al. [7] which consists of FG, PW, and LEO as its key ingredients. GPL coating will then be applied to the created paper samples.

In total, 10 sheets of AHP-GPL were created. Five samples were used for the Cobb test, each measuring 12.7 by 12.7 centimeters in length following the recommended dimensions of the sample by Central Philippine University (CPU) - Packaging Engineering Laboratory. For the tear resistance (TR), five sheets of paper were delivered to the CPU - Packaging Engineering Laboratory for testing.

Preparation of the AHP-GPL. Creating the GPL-coated *Artocarpus heterophyllus* Peel Paper to be used for sample testing involved four steps: formation of paper from AH peels, verification of the AHP, creation of the coating, and the application of GPL coating to the AHP. Paper creation was based on Lothfy et al. [9] with modifications to produce ten sheets of 27 by 22.5 cm AHP. The process began with AH peel drying where the researchers used the laboratory oven from the Philippine Science High School - Western Visayas Campus (PSHS-WVC) heating room, the peels were dried for four hours, at 70° Celsius. The resulting dried peels underwent the soda pulping process, by boiling the peels with 16% (w/w) NaOH for 1 - 2 hours. The resulting pulp was cleaned and dried, then placed in the mold and deckle to form a sheet of paper. The paper in the mold dried after 24 hours. The GPL coating and application were similar to Syahida et al's [7] where a coating made from FG, PW, and LEO is applied to the paper samples.

FTIR Spectroscopy. FTIR Spectroscopy was performed using the Shimadzu IRAffinity-1S at the PSHS Chemistry Instrument Room. The Attenuated Total Reflectance (ATR) accessory of the Shimadzu IRAffinity-1S was used to enhance the surface sensitivity of the machine, leading to more accurate results. Ten scans were performed to ensure the accuracy of the spectra and were set to absorbance, where the peaks were shown going upwards. The range of the scanned spectra was from 700 to 4000 cm^{-1}. The paper composition was confirmed by comparing the spectra to existing literature. [10,13,16].

Water absorption. Five samples of AHP-GPL measuring 12.7 by 12.7 centimeters in length, as recommended by the CPU's Packaging Engineering Laboratory, were used for the measuring of WA. The resulting data were expressed in the standard Cobb Value units: g/m^2.

$$Cobb\,Value = (W_f - W_i) \times 100$$

W_i is the initial weight of the paper after the pouring of water and W_f is the final weight of the paper before the pouring of water.

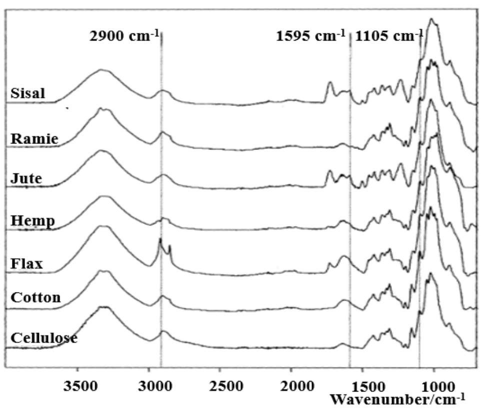

Figure 1. FTIR spectra of cellulose fibers from Garside and Wyeth (2003)

Tear Resistance. After submission to CPU's Packaging Engineering Laboratory, three sheets of the sample material with dimensions of 7.62 by 6.35 centimeters were torn together through a fixed distance using the pendulum of an Elmendorf-type tearing tester in the cross direction, following the standard of ASTM D689. The process was repeated once again in the machine direction. The work done in tearing was measured by the loss of potential energy of the pendulum. The resulting data was measured in grams-force indicating tearing strength (gf). Tear index was then calculated by dividing the average tear resistance of the sample by its grammage.

$$tear\ index\ =\ \frac{average\ tearing\ force\ (gf) \times 9.81}{average\ grammage\ (g/m^2)}$$

Data Analysis. The WA of AHP-GPL was submitted to laboratories for testing and as such, required no further calculations before data analysis. The data was compared to related literature to assess the quality of the two parameters.

Safety Procedure. To ensure safety from possible hazards, proper laboratory procedures were observed by the researchers. Proper laboratory equipment and protective clothing were worn during the handling of chemicals such as NaOH. During the data-gathering process, the Science Research Assistant (SRA) of PSHS-WVC and the research adviser supervised the researchers to ensure their safety.

The proper disposal of the excess materials after the conduct of the data-gathering process was observed. The NaOH used during the soda pulping process was stored in a plastic container as advised by the SRA and was then stored in the Chemistry Laboratory. To avoid clogging the drains of the laboratory, the researchers wait for the palm wax to solidify before scraping and disposal through the garbage gin.

Results and Discussion. - *FTIR Spectroscopy.* The AHP spectra matched the functional groups of cellulose, hemicellulose, and lignin, which are key components of paper. Hydroxyl and methyl bands were found in the single bond area while several tiny sharp peaks were found in the double and triple bond areas. In the fingerprint area, overlapping peaks are widespread and include large ranges and small sharp peaks, most of which are related to the components of the paper. The most significant functional groups of the paper are -OH, -CH, C=O, and C-O. Majority of the peaks in the spectra correspond to cellulose (3036 to 3639, 2800 to 3000,1779 to 1596, 1173 to 840), while the sharp peaks in the single and triple bond region are attributed to lignin's functional groups. The most significant functional groups for paper, according to Munajad et al. [12], are -OH (3700 to 3000 cm^{-1}), -CH (3000 to 2700 cm^{-1}), C=O (1700 cm^{-1}), and C-O (1500 to 900 cm^{-1}). These functional groups coincide with those of cellulose, hemicellulose, and lignin, which are the main components in paper [11,14]. Furthermore, paper contains more cellulose than lignin due to the removal of lignin and other additives during the papermaking process [11] with short, sharp peaks caused by the functional groups of lignin (1499 to 1596; 2397 to 2192; and 2134 to 1870 cm^{-1}) [16, 17].

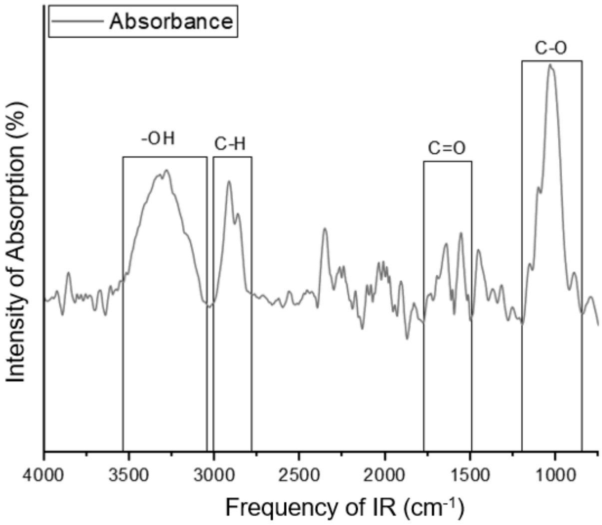

Figure 2. FTIR Spectra of uncoated AHP with the functional groups of paper indicated.

Water Absorption. Laboratory results by CPU - Packaging Engineering Laboratory showed that the Cobb Test of the first three samples had an average water weight of 135 g/m^2. Meanwhile, two of the five submitted samples failed during testing due to leakage. Two of the submitted samples that failed due to leaking may be caused by multiple factors. Lack of equipment in papermaking can lead to an inconsistent thickness around the paper as mechanical papermaking equipment provides a more even pulp distribution [18]. In addition, microscopic holes can occur during the papermaking process, as they cannot be seen by the naked eye.

The water resistance of AHP-GPL is lower than kraft paper, which possesses a water weight of 40 ± 5 g/m^2. A lower water weight value means it has a better water resistance property [7].

Table 1. Cobb test results of AHP-GPL.

Specimen	Weight of water in g/m^2 (Cobb Value)
Sample 1	103
Sample 2	141
Sample 3	161
Sample 4	(Failed)
Sample 5	(Failed)
Std. Deviation	**29.5**
Average	**135 ± 5**

Tear Resistance. The average force required to tear the three samples of AHP-GPL is 87.105 gf. This is converted to millinewtons and divided by the grammage such that the tear index of AHP-GPL is calculated to be 6.117 mN.m^2/g.

Table 2. Tear test results of AHP-GPL

	Cross Direction	Machine Direction
Sample	Tearing Force (g)	Tearing Force (g)
1	80.00	80.00
2	96.00	101.3
3	101.3	64.00
Std. Dev.	**11.10**	**18.73**
Average	**92.44**	**81.77**

This is higher than AHP with no coating, which has a tear index of 0.16698 mN.m^2/g, which might indicate GPL coating's tear resistance-improving properties [9]. However, the increase was not enough to be comparable to kraft paper. Kraft paper is often used for high-tensile packaging applications and has a minimum tear index of 8-10 mN.m^2/g [19, 20, 21]. A factor that can be taken into account is the difference in grammage between the AHP-GPL and kraft paper. The grammage of kraft paper is 70 g/m^2 [20], while the calculated grammage of AHP-GPL is 139 g/m^2. The tear index is also similar among other non-wood fiber papers such as kaun straw, chia, and mustard stalks, which have a tear index of 6.23 mN.m^2/g, 5.9 mN.m^2/g , 5.9 mN.m^2/g respectively [22]. The AHP-GPL has lower mechanical strength than paper made from unbleached kraft pulp, possibly due to the soda pulping process [23]. Although, the TI of AHP-GPL is comparable to other chemical pulps such as softwood sulphite, hardwood sulphite, and hardwood sulphate pulps [15].

Limitations. The tensile strength test cannot be performed due to the lack of an available Universal Testing Machine (UTM). Therefore, tear resistance was the alternative parameter that the researchers tested. Handmade papermaking methods were also utilized, causing inconsistencies in the texture and thickness of the paper created. Furthermore, the unavailability of libraries meant that the assignment of peaks was limited, leading to the reliance on pre-existing literature for FTIR spectroscopy data analysis. Due to the distance of FPRDI, the researchers are unable to conduct the testing of tensile strength in person.

Conclusion. - AHP-GPL packaging paper did not meet the physicochemical standards of other packaging materials and non-wood fiber papers. ATR FTIR spectroscopy showed that AHP contains functional groups present in cellulose, hemicellulose, and lignin, which are major components of paper. However, AHP-GPL had a water absorption Cobb value of 135 g/m^2, which is inferior to other packaging papers. AHP-GPL's tear index was 6.117 mN.m^2/g, lower than kraft paper but higher than AHP without GPL coating, on par with other non-wood fiber papers like kaun straw, chia, and mustard stalks.

Recommendation. - The results of this study may be improved by using a papermaking method that reduces the chance of micro punctures from occurring. The use of equipment dedicated to papermaking can improve the quality of the created *Artocarpus heterophyllus* peel paper, having a more consistent texture and thickness in each area. This can also avoid unnecessary creases and unevenness in the paper. Additional methods during the papermaking process such as pulp beating or adding additives in the pulp can also benefit the properties of the samples. Exploration of different bio-based coatings can also help test the properties of AHP. The tensile strength of the AHP-GPL should also be investigated to obtain a more comprehensive understanding of the mechanical strength of the paper. Additionally, FTIR spectroscopy should also be performed on the coated AHP. Other parameters for packaging paper such as porosity, oil resistance, and burst index should also be tested.

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