

Research Article

The Effect of Eggshell Waste Powder on the Material Properties of the Banana Pulp Papers

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Abstract

Recycling agricultural and food wastes provides a sustainable approach to paper production. This study investigated banana pulp papers with the addition of 10–20 wt.% of eggshell waste powder (ESWP) using fresh and dried banana stems. The addition of ESWP increased paper thickness and weight, reduced moisture content, and enabled rapid water absorption. However, tensile strength and tear index decreased with higher ESWP content due to increased brittleness and the presence of pores and voids. Papers from dried banana stems tended to exhibit higher strength, faster water absorption, and greater moisture retention than those from fresh stems, likely reflecting the effect of thin, continuous fibers in retaining pulp and ESWP. These results demonstrate the potential of using waste to produce environmentally friendly paper products.

Keywords: Banana pulp papers, Eggshell waste powder, Paper characteristics, Tear index, Tensile strength

1 Introduction

Writing, printing, packaging, and other daily applications involve the use of paper. The raw material used in paper production is cellulose fiber, which is derived from both wood and non-wood sources. For the production of environmentally friendly paper, handmade papermaking methods, commonly practiced in local communities, are employed. Agricultural residues such as corn stalks, bagasse, and rice straw [1] serve as raw materials.

Organic waste, e.g., food waste, garden waste, wood waste, and agricultural residues, represents a major component of global waste [2]–[6]. These wastes also continue to increase due to urban expansion, economic progress, and population growth. Several methods of waste disposal, including waste prevention, land-filling, incineration, biological treatment, and recycling [7], [8] have been executed. To ensure environmental sustainability and protect human health, the appropriate waste management should be carefully considered. Banana plantations are commonly found in Southeast Asia (SEA), including Malaysia, Indonesia, Philippines, Vietnam, and Thailand [9]. The harvesting of banana fruit crops

generates large quantities of agricultural waste since banana trees bear fruit only once. After fruiting, the trees need to be cut down for the growth of the new plants around them. Banana tree waste, which contains compounds of nutrition and fiber and can be utilized as organic fertilizer, animal feed, reinforcement materials, and pulp [10]-[13]. Banana pulp or fiber can be extracted from banana stem, which has characteristics suitable for handmade paper [10], [14], [15]. Mote et al., [16] reported that banana fiber, a natural material, is valued for its exceptional mechanical strength. Eggs are a readily available food, low in price, and high in quality protein and nutrition. Thus, eggs are an important ingredient in many dishes and desserts. However, the cooking and baking processes can generate large quantities of eggshell waste. These wastes can be used as fertilizer, an animal feed ingredient, a food additive, an adsorption substance, a filler for composite materials, and others [17]-[20]. Generally, the production of paper in industry uses calcium carbonate, derived from rock and soil resources, as a filler to modify the material properties of paper [21]. The addition of filler for paper making is 10-30%wt filler content [22], [23]. Calcium carbonate is a major component in eggshells



[24], and thus, eggshell waste can be incorporated into paper production, especially in eco-friendly or handmade paper.

The raw materials, composition, and processing methods used in papermaking influence the development of paper quality, such as optical, physical, and mechanical properties, which determine its suitability for different applications. Chandranupap et al., [25] reported that paper produced from recycled paper treated by the deinking process exhibited improved mechanical properties, with the tear index and tensile index increasing by approximately 25% and 18%, respectively. Moreover, differences in natural pulp types, such as eucalyptus, sisal, and pine, had a significant effect on the physical and mechanical properties of paper [26]. For paper production, paper properties can also be enhanced by removing lignin. Alkaline pretreatment, which is one of the main chemical processes used to eliminate lignin, was reported by Areeya et al., [27]. Alkaline agents such as NaOH, KOH, Ca(OH)2, and ammonia are commonly used in this process. Ongsuwan et al., [28] illustrated that the penetration strength, water absorption, and brightness of packaging paper can be improved by the presence of 5 wt.% hydroxyapatite synthesized from bovine bones on the surface of those papers. In addition, other studies have demonstrated the effect of pulp type and eggshell additive on improving paper properties. For instance, Abdullah et al., [19] produced paper using waste paper pulp and eggshell (20-60%wt), with paper containing 20 wt.% eggshell showing maximum stress. Similarly, Fahmy et al., [29] reported that eco-friendly paper made from bagasse pulp mixed with waste eggshells (0–25%wt) can improve grammage, opacity, and brightness, whereas a reduction in mechanical properties, such as tear index and burst index, was observed. In producing eco-friendly paper from organic waste, such as banana trees and eggshells, various factors influence its material properties.

In this work, the influence of eggshell waste powder on the physical and mechanical properties of banana pulp papers made from fresh and dried banana stems was studied. Physical attributes, including morphology, thickness, basis weight, moisture content, and water absorption, were measured. Mechanical performance parameters such as tensile strength, %elongation, and tear index were also determined. The findings and methodology presented here aim to contribute valuable insights toward the advancement of sustainable and eco-friendly materials.

2 Materials and Methods

2.1 Materials

Waste banana stems and eggshell waste were employed as raw materials in this study. The banana stems (40–50 mm in length and \approx 150 mm in diameter) were collected from Ban Mo District in Saraburi Province and Mueang Lopburi District in Lopburi Province, Thailand, and were chopped into small pieces (10-30 mm in width, 60-90 mm in length, and 6-15 mm in thickness). A portion of the chopped banana stems was dried in the sunlight. Fresh and dried banana stems had moisture contents of 90.51 ± 3.77% and $14.88 \pm 2.19\%$, respectively, before being prepared for pulping. Eggshell waste (ESW), another raw material was collected from a bakery in Mueang Lopburi District, Lopburi Province, Thailand. Preliminary pretreatment of ESW involved cleaning with dishwashing liquid and tap water and sun-drying.



Figure 1: Pulping of fresh and dried banana stems.

2.2 Methods

2.2.1 Pulping

Banana pulp, derived from fresh and dried banana stems, was obtained by a chemical process (Figure 1). The cooking liquor was prepared by dissolving NaOH pellets in tap water, consisting of 10% NaOH in 15 liters of tap water. The pot containing the chopped stems and the cooking liquor was heated to boiling and



maintained at a boil for approximately 30 min. For the dried stems, they were soaked in tap water for an hour before heating. After heating, the pulps were cooled in tap water and subsequently washed until the litmus paper indicated neutrality. Approximately 500 g of pulp was then crushed using a kitchen blender (Kashiwa, Thailand) at a maximum speed of 6000 rpm for 45 s. The pulps were stored in a plastic container.

2.2.2 Preparation of eggshell waste powder

Sun-dried eggshells (800 g) were manually crushed and immersed in a 20% NaOH solution for 60 min to remove the inner membrane. They were subsequently washed with tap water until reaching a neutral pH, followed by drying in a hot air oven (Memmert, Germany) at 105 °C for 24 hours. Following this, the dried eggshells (moisture content approximately 0.40 \pm 0.17%) were ground using an electric grain mill grinder (3000W, Zhejiang Boou Electric Appliance Co., LTD., China) to produce eggshell waste powder (ESWP). The ESWP was then sieved using a 0.1 mm mesh (Shaoxing Shangyu District Huafeng Hardware Instrument Co., Ltd., Shaoxing, China) to obtain powder particles with a size $\leq 100 \mu m$, as presented in Figure 2. To prevent moisture absorption, the powder was stored in glass containers.



Figure 2: Preparation of eggshell waste powder (ESWP).

2.2.3 Preparation of paper sheets

Paper sheets were prepared using a traditional papermaking method commonly practiced in local communities. Banana pulp and eggshell waste powder (ESWP) were weighed to obtain a total pulp mass of

1000 g for each sheet. The ESWP content was varied at 0, 10, 15, and 20 wt.%. Both raw materials were combined and blended with 4.5 L of tap water in a box container. Prior to sheet formation, the slurry was manually stirred to ensure uniform dispersion of ESWP. A mold (420 mm \times 594 mm) was then dipped into the pulp slurry. The wet pulp height was estimated at $\sim 1-3$ mm using the mold edge as a visual reference. The sheets were dried at room temperature for approximately 7 days before removal from the mold for property evaluation.

2.2.4 Characterization

The characteristics of the paper samples, including morphology, thickness, moisture content, and water absorption, were investigated. The morphological features of the surface and cross-section were characterized with a scanning electron microscope (SEM, JEOL model JSM-6610 LV, Tokyo, Japan) coupled with energy-dispersive X-ray spectroscopy (EDS, Oxford model INCA350). The bottom surface (in contact with the mold screen) and the top surface (not in contact with the mold screen) were subjected to surface analysis. Paper sheets made from fresh and dried banana stems without ESWP were also characterized by attenuated total reflectance Fourier transform infrared spectroscopy (ATR-FTIR). Three pieces of 10 mm × 10 mm were analyzed, and the spectra were recorded using a Nicolet iS5 spectrometer equipped with an iD7 ATR accessory containing a diamond crystal (Thermo Scientific, USA) in the wavenumber range of 4000–400 cm⁻¹. Paper sheet thicknesses were determined via a digital thickness gauge featuring a maximum measurement range of 12.7 mm and a resolution level of 0.01 mm. Measurement of thickness at eight points per sample was conducted to calculate the average thickness. Five samples, each measuring 100 mm × 100 mm, were evaluated for each condition. These samples were also weighed using a METTLER TOLEDO analytical balance with an accuracy of 0.001 g. The moisture content was determined using a Sartorius MA100 instrument at 105°C with three 50 mm × 50 mm samples per condition. Water absorption was assessed using a water drop test, in which a 50 µl droplet of water was applied to the sample surface, and the time required for complete absorption was recorded. The sample dimensions were the same as those used for moisture content measurement.



2.2.5 Mechanical test

The mechanical performance of the handmade paper was examined by measuring tensile strength, %elongation, and tear index. The tensile test was conducted according to the TAPPI T494-06 standard [30]. Samples with dimensions of 25 mm (width) and 180 mm (gauge length) were prepared, as shown in Figure 3(a). Tests were carried out on a Shimadzu universal testing machine (Kyoto, Japan) equipped with a 1 kN load cell, operating at a constant crosshead speed of 25 mm/min. Ten samples were tested to determine tensile strength and %elongation. Tear index, which is the tear strength corrected for grammage [31], was measured using an Elmendorf tearing tester (ProTear model, Thwing-Albert Instrument Company, Philadelphia, USA) in accordance with the ASTM D689-03 standard. Paper sheets were cut to dimensions of 70 mm × 63 mm as illustrated in Figure 3(b). For paper made from fresh and dried banana stems with and without ESWP, 400 gf and 800 gf pendulums were used, respectively. For each type of paper, five samples were evaluated. The tear index was calculated using Equation (1) [31]:

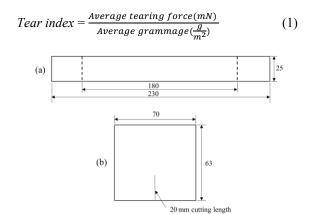


Figure 3: Samples for mechanical testing: (a) tensile specimen, and (b) tear test specimen.

3 Results and Discussion

3.1 Morphology

Figures 4 and 5 illustrate the SEM analysis results of paper sheets made from fresh and dried banana stems containing various amounts of eggshell waste powder (0, 10, 15, and 20 wt.%). The analysis indicates that both types of paper without ESWP (0 wt.%) exhibited a relatively dense surface on the bottom side, whereas pores were observed on the top side. This may be

related to the distribution of pulp fibers during the sheet formation process. In addition, the banana fibers in all paper sheets were arranged in random orientations.

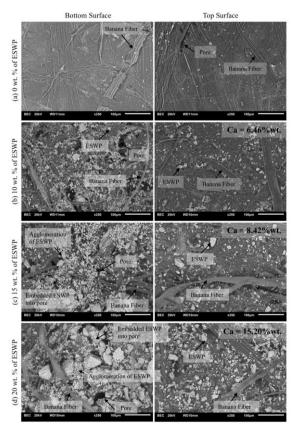


Figure 4: SEM images of the surface of banana pulp papers made from fresh banana stem with eggshell waste powder (ESWP) at concentrations of 0, 10, 15, and 20 wt. % of ESWP.

For the banana pulp papers with ESWP, as shown in Figures 4(b)–(d) and 5(b)–(d), pores could be clearly observed on the bottom surface (the screencontact side) due to the precipitation of ESWP, leading to the inhibition of pulp deposition. Meanwhile, the top surface was relatively less visible compared to paper without ESWP. This can be explained by the lower distribution of ESWP on the top surface and the embedding of ESWP within the pores. Increasing the ESWP content (4.19–17.22 wt.% Ca) resulted in a more extensive and relatively denser distribution of ESWP throughout the paper sheets. However, precipitation, agglomeration, embedment and large ESWP on the subsurface were more evident at higher ESWP contents, partly due to the lack of continuous



stirring in the handmade papermaking method. In addition, the paper made from dried banana stem (Figures 5(a)–(d)) exhibited a more continuous fiber network and thinner fibers compared to that made from fresh banana stems. These characteristics relate to its physical and mechanical properties.

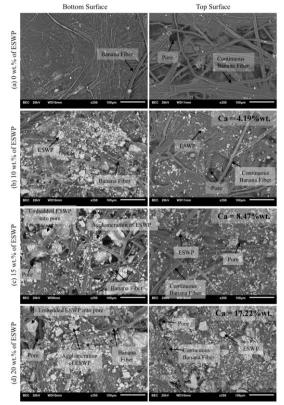


Figure 5: SEM images of the surface of banana pulp papers made from dried banana stem with eggshell waste powder (ESWP) at concentrations of 0, 10, 15, and 20 wt.% of ESWP.

3.2 ATR-FTIR

Comparison of ATR-FTIR spectra of paper made from fresh and dried banana stems without ESWP is shown in Figure 6. Paper sheets produced from 100% pulp of fresh and dried banana stems exhibited similar spectra peaks at ~3300, ~2900, ~1600, ~1400, ~1300, ~1030, and ~550 cm⁻¹. The main functional groups of banana pulp included hydroxyl group (–OH) in cellulose, hemicellulose, and lignin (~3300 cm⁻¹), C–H stretching (~2900 cm⁻¹), the C=O/C=C vibrations (~1600 cm⁻¹), –O–(C=O)–CH3 and –OH (~1300 cm⁻¹), and C–O stretching vibrations (~1030 cm⁻¹) [32]–[34]. Paper made from dried banana stem showed lower

peak intensities, which may reflect partial degradation of hemicellulose and lignin due to the lower moisture content of banana stem (14.88 \pm 2.19%). According to Berrio *et al.* [33], drying at elevated temperature (90 °C) can reduce lignin and other plant-derived components, while peaks at \sim 1400 and \sim 550 cm⁻¹ are likely insignificant or may arise from impurities.

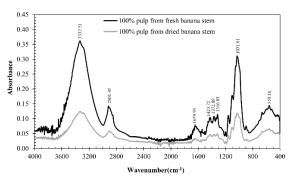


Figure 6: Comparison of FTIR spectra of paper made from fresh and dried banana stems.

3.3 Thickness, weight, moisture content, and water absorption

Table 1 shows the thickness, weight, moisture content, and water absorption of banana pulp papers with and without eggshell waste powder (ESWP). Both thickness and weight generally increased with higher ESWP content. At 20 wt.% of ESWP, the papers reached their highest values (thickness: 1.26-1.49 mm; weight: 673-681 g/m²), likely due to ESWP filling the pores, adhering to banana fibers, and agglomerating within the sheets (Figures 4 (b)–(d) and 5(b)-(d)). Thickness and weight variations of paper with high ESWP were also observed, probably caused by non-uniform powder distribution. Papers made from dried banana stems exhibited higher thickness and weight than those from fresh stems, because of the presence of a more continuous and thinner network fiber (Figure 5). They could act as filters, better capturing smaller ESWP particles and fine banana fibers (Figure 7) during the papermaking process. High variations in the weight of paper with ESWP can be found due to the random orientation of thin fibers, leading to uneven deposition of pulp and ESWP. However, for practical applications such as paperboard, packaging, and printing, paper weight should remain below 600 g/m². Typically, general paper ranges from 60 to 300 g/m², while specialty boards for packaging (e.g., postcard board, boxboard) range from 150 to 500 g/m² [35].



Table 1: Thickness, weight, moisture content, and water absorption of banana pulp papers with and without eggshell waste powder.

Туре	Thickness (mm)	Weight (g/m²or grammage)	Moisture Content (%)	Water Absorption (s)
100% pulp from fresh banana stem	0.47 ± 0.06	125 ± 19	5.88 ± 0.48	14.65 ± 2.75
90% pulp from fresh banana stem + 10%ESWP	0.87 ± 0.17	333 ± 69	1.32 ± 0.34	4.55 ± 0.65
85% pulp from fresh banana stem + 15%ESWP	0.86 ± 0.23	363 ± 150	1.43 ± 0.25	6.33 ± 1.53
80% pulp from fresh banana stem + 20%ESWP	1.26 ± 0.32	681 ± 206	0.95 ± 0.11	2.16 ± 0.34
100%pulp from dried banana stem	0.81 ± 0.11	242 ± 39	7.37 ± 0.46	10.75 ± 3.53
90% pulp from dried banana stem + 10%ESWP	0.95 ± 0.11	352 ± 53	3.31 ± 0.55	3.25 ± 0.40
85% pulp from dried banana stem + 15%ESWP	1.48 ± 0.22	653 ± 104	2.43 ± 0.23	4.84 ± 1.03
80% pulp from dried banana stem + 20%ESWP	1.49 ± 0.18	673 ± 146	2.25 ± 0.20	3.37 ± 0.67

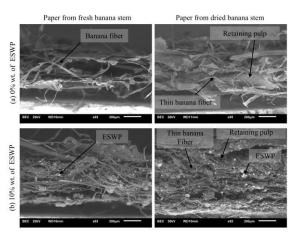


Figure 7: Cross-sectional images of banana pulp papers made from fresh and dried banana stems.

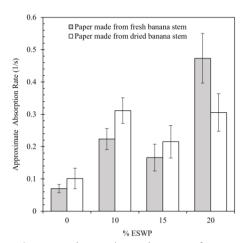


Figure 8: Approximate absorption rate of paper made from fresh banana stem.

The addition of ESWP reduced the moisture of paper sheets, due to the low water content of the eggshell powder (less than 1%) and the formation of pores within the paper. This property tends to decrease slightly as the paper contains more eggshell powder, likely because the presence of additional pores

increased the opportunity for water evaporation, as also suggested by Gonçalves *et al.* [36], since porous materials can accelerate the evaporation rate of liquids. Furthermore, higher variability is observed in the paper without ESWP, likely due to fewer pores, which may limit water evaporation.

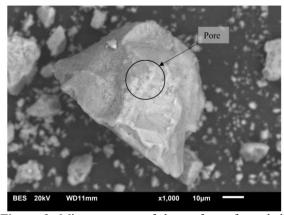


Figure 9: Microstructure of the surface of eggshell waste powder.

Similarly, the water absorption of papers containing ESWP (0.1-0.5 s⁻¹ for approximate absorption rate in Figure 8) was faster than that of the paper sheets without ESWP (<0.1 s⁻¹; Figure 8). This enhancement can be attributed to the porous nature of ESWP particles (Figure 9) and the interfacial gaps between ESWP and pulp fibers (Figure 10), consistent with the findings of Farahana et al. [37]. A notable observation was that the paper made from fresh banana pulp with 20% ESWP exhibited a relatively high approximate water absorption rate compared to other compositions. This is likely due to the relatively dense distribution of ESWP throughout the sheet and the aggregation of larger particles (Figure 4(d)), which created additional pores and promoted faster water penetration.



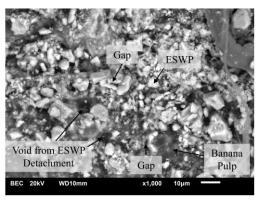
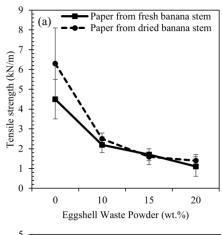


Figure 10: SEM images of gaps, voids and embedded ESWP.

Additionally, the use of dried banana stems for producing banana pulp paper also promoted moisture by more than 40% and accelerated water absorption by more than 20% compared to paper made from fresh banana stems. This behavior may be influenced by the thinner and smoother dried banana fibers (Figure 5). These thin and smooth fibers provide a larger specific surface area and form finer capillary channels, which enhance water penetration and increase moisture retention in the paper structure [36], [38], [39].

3.4 Mechanical properties

Figure 11 shows the influence of ESWP on the tensile strength and percentage elongation of banana pulp papers. Papers without ESWP exhibited the maximum tensile strength (≈4 kN/m for paper made from fresh banana stems and ≈6 kN/m for papers made from dried banana stems). The strength appeared to decrease with increasing ESWP content, and a similar trend was observed for the ductility of paper (Figure 11(b)). The failure of all samples was brittle fracture, as shown in Figure 12. This behavior results from discontinuous fibers and low moisture content of the papers. The addition of ESWP creates voids and gaps in the paper sheet, which can serve as initiation points for failure. A previous study by Hubbe and Gill [40] reported that the rigid fillers, such as ESWP, could not enhance the paper strength as cellulosic fibers. The low moisture content of the paper containing ESWP (less than 4% as shown in Table 1) is expected to reduce flexibility, resulting in lower %elongation. Papers with low moisture content tend to be hard and brittle, making them prone to web breaks, poor printing performance, and cracking when folded. Nevertheless, among the papers with ESWP, those containing 10 wt.% ESWP displayed higher tensile strength compared to papers with other ESWP proportions, which is likely influenced by the fibers.



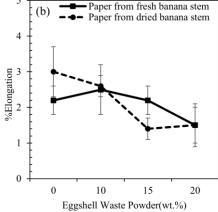


Figure 11: Mechanical properties of banana pulp paper: (a) Tensile strength, and (b) %Elongation.

For paper made from 100% pulp of dried banana stems, the presence of continuous thin fibers relatively enhanced tensile strength. As reported by Berrio *et al.* [33], thinner fibers exhibited higher tensile strength, which was attributed to a more compact internal structure. In addition, the relatively low lignin content indicated by the FTIR spectra (Figure 6) may further promote tensile strength by allowing stronger hydrogen bonding between fibers. However, high fluctuations in tensile strength and %elongation were observed in all samples. There were no significant differences in tensile properties between papers made from fresh and dried stems.

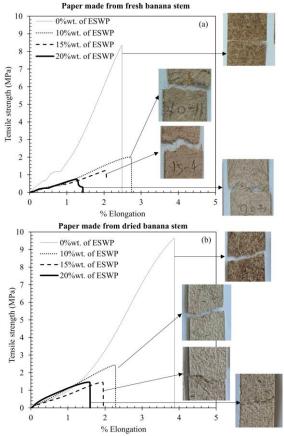


Figure 12: Fracture of tensile sample: (a) Paper made from fresh banana stem, and (b) Paper made from dried banana stem.

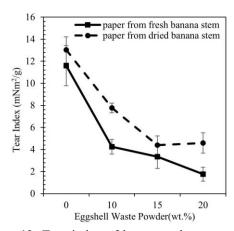


Figure 13: Tear index of banana pulp papers made from fresh and dried banana stem with eggshell waste powder (ESWP) at concentrations of 0%,10%, 15% and 20% wt. of ESWP.

The tear index of banana pulp papers made from fresh and dried banana stems with 0, 10, 15, and 20 wt.% ESWP is shown in Figure 13. The results indicate that the tear index decreases as ESWP content increases. It should be noted that a high amount of ESWP may weaken the fiber structure as reflected in the tensile strength results. Furthermore, the use of dried banana stems as the primary material in paper manufacturing led to an enhanced tear index. This effect can be attributed to their higher moisture content (\approx 7% for paper without ESWP and \approx 2.5% for paper with ESWP), which may reduce the brittleness of the paper pulp and consequently enhance tear strength. This means that the moisture content has a greater effect on the tear index than tensile strength.

4 Conclusions

The addition of 10–20 wt.% of eggshell waste powder (ESWP) into banana pulp papers created structures with embedded particles, pores, and voids. Paper thickness increased by 50-260% and weight by 140-280%. At 15-20 wt.% of ESWP, the thickness exceeded 1 mm and the weight was greater than 600 g/m², which are unsuitable for common applications. Papers containing ESWP exhibited reduced moisture content (<45%) and rapid water absorption, completed within 7 s (≈ 0.5 s⁻¹). The highest approximate absorption rate for papers from fresh banana stems was observed at 20 wt% ESWP, attributable to increased pores and random fiber arrangements. However, tensile strength and tear index were not enhanced, with tensile strength decreasing by approximately 50% due to increased brittleness and pores, which promoted early failure. Papers produced from dried banana stems showed relatively lower hydroxyl and lignin levels and exhibited thinner, more continuous fibers compared to papers from fresh banana stems. Their thickness increased by 100–175% and weight by 100-180% due to the networked fiber structure that retained both pulp and ESWP. These papers also accumulated 180-250% more moisture and absorbed water about 70% faster. Maximum tensile strength (≈6 kN/m) was observed only in driedstem papers without ESWP, while tear strength improved with dried stems at all ESWP loadings, likely influenced by a higher moisture content $(\sim 1.5\%)$. These results demonstrate the potential of combining banana and eggshell waste to produce ecofriendly paper materials with enhanced bulk and absorption properties, suitable for applications where mechanical strength is less critical.



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Author Contributions

The author is solely responsible for the conception, design, data collection, analysis, and writing of this manuscript.

Conflicts of Interest

The authors declare no conflict of interest.

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