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Research article

Utilization of pineapple leaf fiber mixed with banana or cattail stem fibers and their paper physical properties for application in packaging

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Abstract

Importance of the work: Recent shortages of wood and non-wood fibers for papermaking have encouraged research on new cellulosic fiber sources from agricultural wastes.

Objectives: To utilize pineapple leaf, banana pseudostem and cattail stem as agricultural wastes in Thailand for papermaking.

Materials & Methods: A completely randomized design was applied for each raw material. Chemical compositions and fiber morphologies were analyzed to determine the best treatments for all raw materials. Then, the optimum conditions for pulping and bleaching were assessed based on characterization of the yield content and pulp brightness.

Results: The results showed that 30% sodium hydroxide produced the best pulping conditions, while 40% hydrogen peroxide produced the highest brightness. Then, bleached pineapple pulp was mixed with banana or cattail pulps at various ratios to study the effect of the pineapple pulp with other pulp blends on their characterization of blended paper. The appropriate properties were identified as 70% pineapple pulp and 30% banana or cattail pulps. Then, mixtures of pineapple-banana or pineapple-cattail pulps at a ratio of 70:30 were formed and coated with 0.6% (volume per volume, v/v) of chitosan solution and 2.0% (v/v) of water-resistant agent. It was found that the mechanical and water-resistant properties of pineapple-banana paper were improved by coating with 0.6% chitosan solution and 2.0% water-resistant agent.

Main finding: Paper made from a pineapple-banana mix showed promise as packaging paper, with good mechanical and water-resistant properties.

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Introduction

Packaging materials for the food processing industry include paper, plastic, glass, aluminum and wood that are used either singly or in combination depending on requirements (Jones and Comfort, 2017). The paper packaging market, consisting of paper and paperboard, was estimated to be worth USD 342.77 billion in 2019, with a compound annual growth rate of 4.5% forecast from 2020 to 2025 (Industry Arc, 2019). Due to global environmental concerns, paper is predominantly used at primary (in direct contact with food products) and secondary levels (Khwaldia et al., 2010).

The raw materials used in papermaking can be classified into three groups: wood, non-wood and recycled waste paper. Resources of natural wood are declining and the development of non-wood raw materials for papermaking has recently attracted increased attention; however, this uses a lot of energy and water (Triastuti, 2020). The advantages of non-wood plants as alternative fiber resources include fast growth and reduced lignin content, requiring lower energy and chemical reagents during the pulping process (Rodríguez et al., 2008). However, sources of non-wood pulp are limited and non-wood fibers from different sources are usually mixed to obtain good quality fibrous material to enhance distinctive paper properties (Sridach and Paladsongkhram, 2014). Non-wood plant short fibers, such as rice straws, corn stalks, bagasse, cattail stems and banana pseudostems, have been used as materials for paper packaging (Laftah et al., 2015; Chollakup et al., 2020, 2021; Vaithanomsat et al., 2021). Non-wood plants currently used to make paper packaging include pineapple leaf fibers, oil palm empty fruit branches, paper mulberry, jute and kenaf. The shorter fiber is used as a filler to fill in the paper's pore and improve the tensile strength.

Pineapple leaves left in the field as a waste raw material are used to produce natural fiber. This waste after fruit harvest in Thailand is estimated at 7.68 Mt per year, and the leaves could produce fibers amounting to 153.60–230.40 kt in 2022 (Apipatpaph et al., 2022). Banana pseudostem is one of the agricultural short fiber sources produced from a fruit that is abundant in many tropical areas, with the global production of banana fiber being around 100,000 t/yr (Subagyo and Chafidz, 2018). Cattail is an easy-to-grow water plant that can be found throughout wetlands and the stems are a source of lignocellulosic fibers that show potential as a fiber for paper pulp (Sridach and Paladsongkhram, 2014). Mechanical and other specific properties of mixed pulp paper, such as water resistance, can be improved using chemical reagents (Zhang et al., 2014).

Chitosan is normally obtained from the alkaline deacetylation of chitin extracted from the exoskeletons of crustaceans, arthropods and mollusks (Arasukumar et al., 2019). It is used in many applications, such as in the textile, paper, packaging, food, pharmaceutical and tissue engineering industries but also in commercial packaging because of its antimicrobial properties (Gatto et al., 2019; Tanpichai et al., 2020). Incorporation of chitosan in paper pulp by sizing or coating creates strong bonds between the molecular structures of the cellulose and chitosan that improve the mechanical properties of the paper; furthermore, the addition of chitosan improves hydrophobic properties by decreasing water absorption, increasing wetting time and reducing the air permeability of the paper (Laleg and Pikulik, 1992; Zhang et al., 2014).

Other biomaterials, such as polysaccharides (sodium alginate and gellan gum), proteins, lipids, biodegradable polyesters (poly-hydroxyalkanoate and polylactic acid) have also attracted interest as paper or paperboard coatings (Iewkittayakorn et al., 2020; Vaithanomsat et al., 2021). Vaithanomsat et al. (2021) concluded that biosynthesized poly(3-hydroxybutyrate)-coated pineapple paper prevented water drop penetration and underwent biodegradation. However, this coated paper has not been used commercially in industrial paper applications. Perfluorooctanoic acid (PFOA)-free fluorocarbon that resists grease and water has long been used to treat paper wraps for burgers and other foods and also in water repellent finishes on fabrics (Qi et al., 2002). This nonflammable fluorocarbon based on C-6 chemistry has been applied as a durable water and oil repellent agent for both synthetic and cellulosic fibers (Smet et al., 2015).

The current study utilized pineapple leaf, banana pseudostem and cattail stem to produce paper and packaging bags with optimum pulp mixtures. First, the chemical compositions and fiber morphology of pineapple leaf, banana pseudostem and cattail stem were analyzed to determine the best treatments for their raw materials. Then, the optimum conditions for preparing and bleaching the three pulp sources were assessed based on characterization of the yield content and pulp whiteness.

Materials and Methods

Materials and characterization

Agricultural waste consisting of pineapple leaves, banana pseudostems (collected after harvesting the main crop) and cattail stems as a weed were selected as the raw materials (Fig. 1) and prepared for fiber extraction using a decorticating machine. The pineapple (*Ananas comosus*, Smooth cayenne) leaves were collected after last fruit harvesting at age 3.5 yr, as determined in Ratchaburi, Thailand. The banana (*Musa ABB cv*. Kluai "Namwa") pseudostems were collected at age 2–3 yr, as determined in Petchaburi, Thailand. The cattail (*Typha angustifolia*) stems were collected as weeds after growing for 2 yr in a field near Kasetsart University, Bangkok, Thailand. After the collection times mentioned for the current study, the residues were usually left on the ground until the next crop. The collected raw materials were dried at 75 °C for 16 h until moisture content was 9–12% on a dried basis; then, they were cut into 1–2 cm lengths. Plant parts and fibers of pineapple leaf, banana pseudostem and cattail stem were extracted using a decorticating machine (Fig. 1). Chitosan (low molecular weight grade with 99% degree of deacetylation) was purchased from Marine Bio Resources, Co., Ltd (Thailand) and coated on the paper surface at about 10 g/m^2 to improve the mechanical properties. A water- and oil-repellent agent, namely perfluorooctanoic acid-free fluorocarbon (PFOA-free fluorocarbon) was purchased from Star Tech Chemical Co., Ltd. (Bangkok, Thailand).

The chemical compositions of the three pulps were analyzed based on standard TAPPI methods for alpha-cellulose (TAPPI T 203 cm-22, TAPPI, 2009b), holocellulose (browning, 1967), extractives (T 204 cm-17, TAPPI, 2009g), lignin content (TAPPI T222 om-21, TAPPI, 2009a) and ash (TAPPI T211 om-22, TAPPI, 2009c).

The fiber morphology of each pulp was determined following the method of Franklin (1945). The pulps were dispersed in glacial acetic acid and hydrogen peroxide at 60 °C for 48 h. The fibers were viewed under a light microscope (LM750; Leica; Wetzlar, Germany) to measure a single fiber length and diameter to calculate the slenderness ratio (fiber length: fiber diameter).

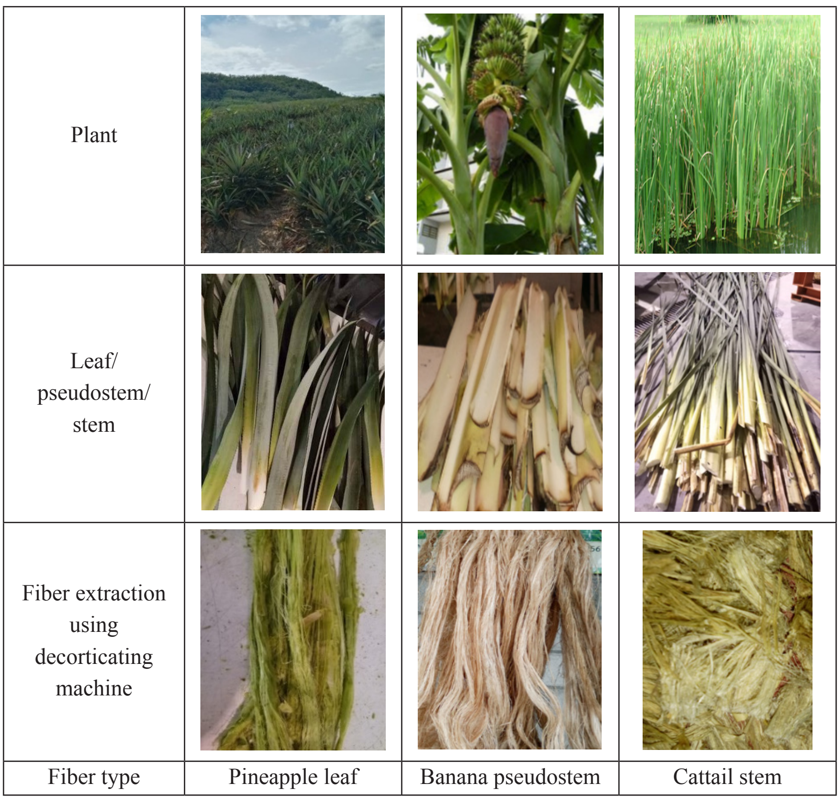

Fig. 1 Pineapple leaf, banana pseudostem and cattail stem showing their extracted fibers following decortication

Preparation of pineapple, banana and cattail pulps

The samples of pineapple leaf, banana pseudostem and cattail stem were treated following the modified method of Chollakup et al. (2021). The raw materials were boiled separately with 10%, 20%, 30% or 40% sodium hydroxide (NaOH) solutions for 3 h at a liquor-to-non-wood ratio of 20:1. Then, the obtained pulps were washed with distilled water to remove the remaining alkali until the pH was equal to 7. The percentage of yield after treatment was calculated. The brightness value of each pulp was measured using a portable colorimeter (Miniscan EZ 4500L; Hunter Lab; Reston, VA USA).

Bleaching process for pineapple, banana and cattail pulps

After alkaline treatment, each pulp was bleached with 10%, 20%, 30% or 40% hydrogen peroxide (H_2O_2) solution. The oven-dried weights of the pulps were mixed with 2% sodium silicate (Na_2SiO_3) and 2% magnesium sulfate ($MgSO_4$) as catalysts for 3 h at 100 °C at a liquor-to-non-wood ratio of 20:1. After that, the bleached pulps were thoroughly washed using distilled water until reaching pH 7, with the associated paper formed at 70 ± 5 g/m^2. The brightness of the papers was measured using a MiniScan EZ instrument (Miniscan EZ 4500L; Hunter Lab; Reston, VA, USA).

Paper preparation from pineapple-banana and pineapple-cattail mixtures

The main components was pineapple fiber (60%, 70% or 80%) filled up with the shorter fiber of the banana or cattail pulp (40%, 30% or 20%) by oven-dried weight. In addition 100% pineapple pulp was formed into paper to compare with the other combinations. The mixed pulps were prepared into hand sheets following the TAPPI T 205 sp-18 method (TAPPI, 2009e). After mixing the samples in a disintegrator, each mixture ratio was formed into paper sheets at 70 ± 5 g/m^2. The mechanical properties of the papers were analyzed following the TAPPI methods for basis weight (TAPPI T410 om-19 TAPPI, 2009f), tensile strength (TAPPI T494 om-22 TAPPI, 2009i), tearing strength (TAPPI T414 om-21, TAPPI, 2009g), bursting strength (TAPPI T403 om-15 TAPPI, 2009d) and folding endurance (TAPPI T423 cm-21 TAPPI, 2007).

The basis weight (g/m^2) was measured following the standard of TAPPI T410 om-19 (TAPPI, 2009f), referring to the weight of the paper per one unit of paper area under specific relative humidity (50% at 23 °C).

Tensile properties, as presented by the tensile index of the paper samples, were analyzed in accordance with TAPPI T494 om-22 (TAPPI, 2009i) using a Schopper tensile tester (Kumagai Riki Kogyo Co., Ltd., Tokyo, Japan). The tensile testing was performed at a strain rate of 25 ± 5 mm/min and a clamp distance of 100 mm. The breaking force value (N) was recorded and used to determine the tensile index (in newtons meter per gram) according to Equation 1:

$$\text{Tensile index} = [653.8 \times \text{Breaking force}] / \text{Basis weight} \quad (1)$$

The tearing strength values of the paper samples were analyzed using a tearing strength tester (Kumagai Riki; Kogyo Co., Ltd./ Tokyo, Japan) according to TAPPI T 414 om-21 (Elmendorf method, see TAPPI, 2009g). The test specimens were prepared in a rectangular size of 6.3 cm × 10 cm. The tear resistance force (N) was recorded and then the tear index was calculated according to Equation 2:

$$\text{Tear index (mN·m}^2\text{/g)} = [9.807 \times \text{tear strength}]/\text{basis weight} \quad (2)$$

Burst index was determined using a Mullen bursting strength tester (Kumagai Riki

Kogyo Co. Ltd.; Tokyo, Japan) according to TAPPI T403 om-15 (TAPPI, 2009d). A rectangular paper sample (12.5 cm × 12.5 cm in size) was prepared. A pressure gauge on the instrument provided a measure of the bursting pressure needed to rupture the paper. Bursting strength was reported in kilopascals (kPa) and then the burst index was calculated using following Equation 3:

$$\text{Burst index (kPa·m}^2\text{/g)} = \text{burst strength/basis weight} \quad (3)$$

The folding endurance test was measured following the standard of TAPPI T423 cm-21 (TAPPI, 2007) using an MIT folding endurance tester (Kumagai Riki Kogyo Co. Ltd.; Tokyo, Japan). The folding endurance or the number of double folds was defined as the number of repeated forward and backward folds the specimen could withstand under 1 kg tension before breaking.

Comparing different paper weights and coating treatments

The samples of pineapple-banana and pineapple-cattail paper at optimum ratios were prepared at 150 and 190 g/m^2 and then coated with chitosan and PFOA-free fluorocarbon using rod coater No. 20 to obtain coated paper with a basis weight of 160 g/m^2 or 200 g/m^2. Chitosan solution at 0.6% concentration was selected from previous study (data not shown), while PFOA-free fluorocarbon at 2% concentration was selected as the minimum amount suggested in the leaflet sheet. The mechanical properties and water resistance of the prepared papers were analyzed.

Statistical analysis

Data were subjected to one-way analysis of variance in a completely randomized design (CRD) for each raw material, with statistical significance set at $p < 0.05$ in the SPSS 17.0 for Windows software (SPSS Inc; Chicago, IL, USA). Duncan's new multiple range test was used to determine significant differences between treatments. Three replications were used to determine each studied treatment. For each characterization of pulp and paper, at least five repeated observations were used to determine each property. Each studied treatment of the CRD design in each factor was the optimum condition of alkaline treatment for each pulp and the optimum condition of bleaching treatment for each pulp. Then, the optimum pulp mixing ratio and all paper properties prepared from mixtures of pineapple-banana pulps were compared to pineapple-cattail pulps.

Results and Discussion

Chemical compositions of pineapple leaf, banana pseudostem and cattail stem

Table 1 shows the chemical compositions of pineapple leaf, banana pseudostem and cattail stem. The pineapple leaf had the lowest extractive content or amount of non-volatile, solvent-soluble material. Higher extractive contents are associated with lower crystallinity and a lower cellulose content or smaller cellulose crystallite size (Blanco et al., 2018). Alpha-cellulose, hemi-cellulose and holocellulose were also detected, with the highest contents in the pineapple leaf. The banana pseudostem had higher lignin and ash contents than the cattail stem and pineapple leaf, which needed to be removed from the fibers. The banana pseudostem contained mostly potassium and calcium compounds that caused the high ash content. The cellulose-to-lignin ratios of the three plant sources were greater than 2, suggesting normal alkaline pulping (Sridach and Paladsongkhram, 2014).

Fiber morphology represented as the slenderness ratio was calculated from the ratio of fiber length to fiber diameter. This affects paper strength (Zobel and van Bujienen, 1989). Generally, the slenderness ratio should be more than 75 to optimize paper tearing strength and a longer fiber length impacts the tearing resistance of paper (Oluwadare and Ashimiyu, 2007). Paper made from the pineapple leaf fiber had the highest slenderness ratio and was more resistant to tearing, while the slenderness ratio of the banana fiber was below 75 and not suitable for forming paper with a high tearing strength (Table 2). However, the banana fiber could be used as filler for the pineapple paper. Therefore, pineapple fiber was selected as the main paper component, with banana or cattail fibers used as fillers.

Table 2 Fiber length, fiber diameter and slenderness ratio of single fibers from pineapple leaf, banana pseudostem and cattail stem

Source of single fiber	Fiber length (mm)	Fiber diameter (μm)	Slenderness ratio
Pineapple leaf	2.45±0.12[a]	10.58±0.55[b]	231.57±21.82[a]
Banana pseudo stem	1.55±0.07[b]	22.20±1.59[a]	70.45± 4.38[b]
Cattail stem	0.76±0.04[c]	9.62±0.50[c]	79.00± 8.72[b]

Mean ± SD in the same column superscripted with different lowercase letters are significantly ($p < 0.05$) different.

Table 1 Chemical compositions of pulp sources from pineapple leaf, banana pseudostem and cattail stem

Source	Chemical component					
	Holo cellulose	Alpha- cellulose	Hemi-cellulose	Extractives	Lignin	Ash
Pineapple leaf	77.66±3.88[a]	51.81±2.59[a]	25.62±1.28[a]	8.80±0.44[b]	9.93±0.49[b]	3.05±0.15[b]
Banana pseudostem	63.47±3.17[b]	49.86±2.49[a]	15.56±0.78[b]	10.88±0.54[a]	17.43±0.87[a]	13.47±0.67[a]
Cattail stem	57.63±3.03[c]	44.02±2.20[b]	19.62±0.98[b]	10.61±0.53[a]	10.10±0.50[b]	2.10±0.10[c]

Mean ± SD in the same column superscripted with different lowercase letters are significantly ($p < 0.05$) different.

Alkaline pulping and bleaching process for pineapple, banana and cattail pulps

Alkaline treatment at higher concentrations of NaOH gave decreased yields for all pulps, as shown in Fig. 2A. Impurities were extractive hemicellulose and lignin contained in the pulp. The pulp appearance was determined for each pulp treatment for 10–20% NaOH which still meant there was incomplete removal of other impurities, as shown by the pulp appearance (Fig. 2A) that was different to the treatments with 30–40% NaOH. A higher alkaline concentration removed the impurities and also extracted and removed some cellulose from the fibers (Triastuti, 2020). The banana fiber produced the highest yield after treatment at all NaOH concentrations, while all fibers treated with 10–20% NaOH showed incomplete removal of other impurities, as shown by the pulp appearance (Fig. 2A). In addition the fibers were lumped together. All pulps were completely separated from fiber using 30% NaOH solution and this concentration was selected for pulping the three raw materials, producing pulp yields of pineapple, banana and cattail of 37.33%, 56.92% and 35.66%, respectively. After bleaching, each pulp at concentrations of 10–40% H_2O_2 solution was formed into paper at 70 ± 5 g/m^2 as the general basis weight of paper. The brightness of all papers increased with increasing H_2O_2 concentration (Fig. 2B); 40% H_2O_2 was selected for bleaching the three pulps to characterize the mechanical properties of the paper.

Characterization of pineapple-banana paper and pineapple-cattail paper

The mechanical properties of the paper samples formed by mixing pineapple-banana or pineapple-cattail pulps at different ratios are shown in Fig. 3. At the same ratios of each pulp mixture, the folding resistance index, tensile index, tear index and burst index of the pineapple-banana paper samples were higher than for the pineapple-cattail paper at a mixture of pineapple pulp greater than 70%. This was due to the thicker and longer fibers of the banana pulp compared to the cattail pulp being better dispersed in the network of the finer pineapple fibers. Blending of long and short fiber pulps enhances the strength and formation of the paper (Retulainen et al., 1998). The cattail pulp had a similar fiber diameter but a shorter fiber length than the pineapple pulp fibers, with the network of interfacial fiber bonding being discontinuous and weak, resulting in reduced paper strength compared to the pineapple-banana paper samples.

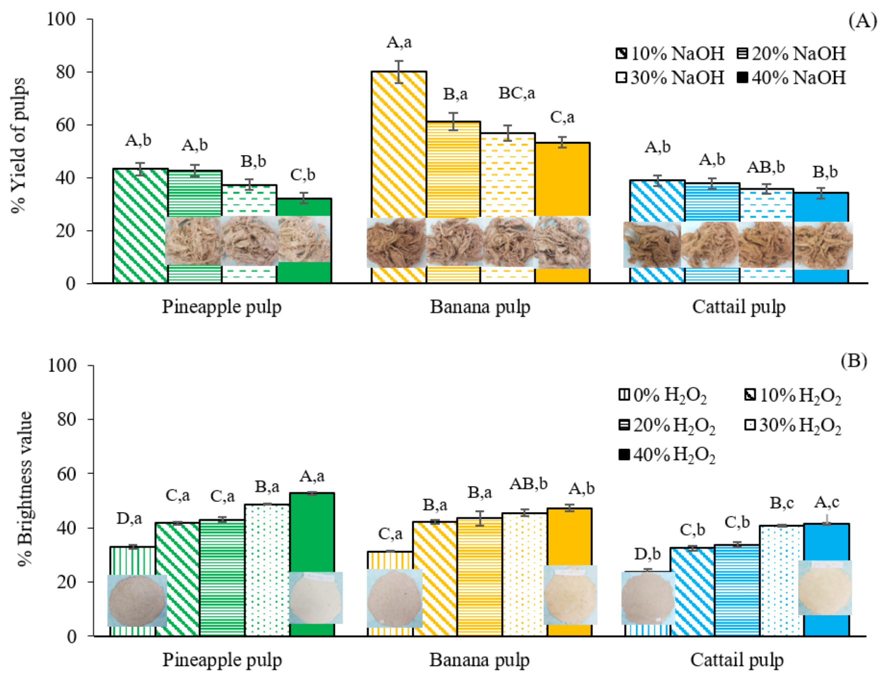

Fig. 2 Percentage yield (A) of each pulp after treatment with different NaOH concentration with pulp appearance; and percentage brightness (B) of each pulp after bleaching with different concentrations of H_2O_2, where different capital letters indicate significant ($p < 0.05$) difference among means within each pulp source;, different lowercase letters at same NaOH (or H_2O_2) concentrations indicate significant ($p < 0.05$) differences

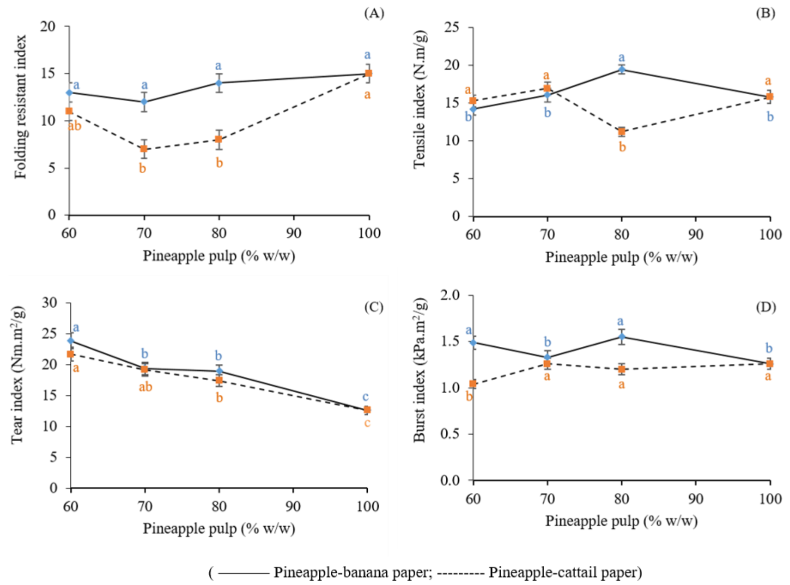

Fig. 3 Mechanical properties of different pineapple-banana and pineapple-cattail paper ratios at 70 g/m^2: (A) folding resistance index; (B) tensile index; (C) tear index; (D) burst index, where different lowercase letters in same pulp source indicate significant ($p \leq 0.05$); difference w/w = weight per weight

Inclusion of the pineapple pulp in the mixture increased the folding endurance (double fold) and the tensile index of the paper because the pineapple pulp had a high slenderness ratio compared to the other pulps. A positive correlation exists between slenderness ratio and folding endurance (Ona et al., 2001; Sarogoro and Emerhi, 2015); in the current study, adding the pineapple fiber increased the folding resistance index. The tensile index results showed that the pineapple pulp blended with the banana pulp better than with the cattail pulp because the pineapple-banana fibers were generally longer and more flexible than the pineapple-cattail fibers. Thus the fiber length affected the tensile strength of the paper.

Fig. 3 shows that tear indices of the pulp blends decreased with increasing pineapple pulp content. The tear index depended on the fiber length, total number of fibers and the fiber-to-fiber bonds in the pulp sheets. The paper was formed by hand and the filler as banana or cattail fiber resulted in fiber-to-fiber bonding in the pores between networks of pineapple fiber. Mixing of 40% thicker fibers produced the highest paper tear index.

In general, the burst strength is highly correlated to the tensile strength (El-Hosseiny and Anderson, 1999). Increasing the pineapple fiber content in the hand-formed sheets affected the burst index (Fig. 3C), which tended to increase with longer and fine fiber of pineapple fiber content.

Banana pulp was a more suitable filler in pineapple paper than cattail pulp. The ratio of 70:30 of pineapple-to-banana or pineapple-to-cattail was selected for the next study because this gave appropriate mechanical properties for both the banana and cattail pulp mixtures. A ratio of 60% pineapple pulp mixed with 40% of the other two pulps had decreased tensile and bursting indices of the papers. Notably, the paper formed from a mixture of two pulps at a basis weight of 70 g/m^2 was used as the general basis weight to determine which ratio provided the best properties of paper for each mixture. In the next step, paper samples at basis weights of 160 g/m^2 and 200 g/m^2 (after coating), which are the basis weights used for bag, were prepared from the optimum ratio and combination for comparison.

Mechanical properties of banana/pineapple paper and cattail/pineapple paper after coating with 0.6% chitosan and 2.0% perfluorooctanoic acid-free fluorocarbon at basis weight of 160 g/m^2 and 200 g/m^2

The pineapple-to-banana paper and the pineapple-to-cattail paper at the ratio of 70:30 selected from the previous section were prepared at 150 g/m^2 and 190 g/m^2. The papers were coated with 0.6% chitosan and 2.0% PFOA-free fluorocarbon as the optimum concentrations determined for water-resistant properties in a preliminary study. The coated weight was about 10 ± 5 g/m^2. The coated weight was controlled by using the coating agent concentration and rod coater number. Commercial bags are mostly coated with PFOA-free fluorocarbon to improve water and oil resistance (Hubbe and Pruszynski, 2020). In fact, the coating weight is an important factor affecting the mechanical properties; increasing the coating weight led to increased tensile strength and tearing resistance of coated papers (Khwaldia, 2013)

Fig. 4A shows the mechanical properties of the pineapple and-banana paper and the pineapple and cattail paper after surface coating. The pineapple-banana paper (160 g/m^2, after coating) had higher tensile strength, tearing strength and bursting index than the pineapple-cattail paper, while the pineapple-cattail paper had better folding resistance than the pineapple-banana paper. At both paper grammages, the water-resistance value of the pineapple-banana paper was very similar to that of the pineapple-cattail paper. However, the tear index and bursting index of the pineapple-banana papers were lower than for the pineapple-cattail papers. The pineapple-banana paper at 200 g/m^2 (Fig. 4B) had better mechanical properties than the pineapple-cattail paper. These results suggested that the pineapple-banana mixed papers showed promise as packaging paper, with good mechanical and water-resistant properties.

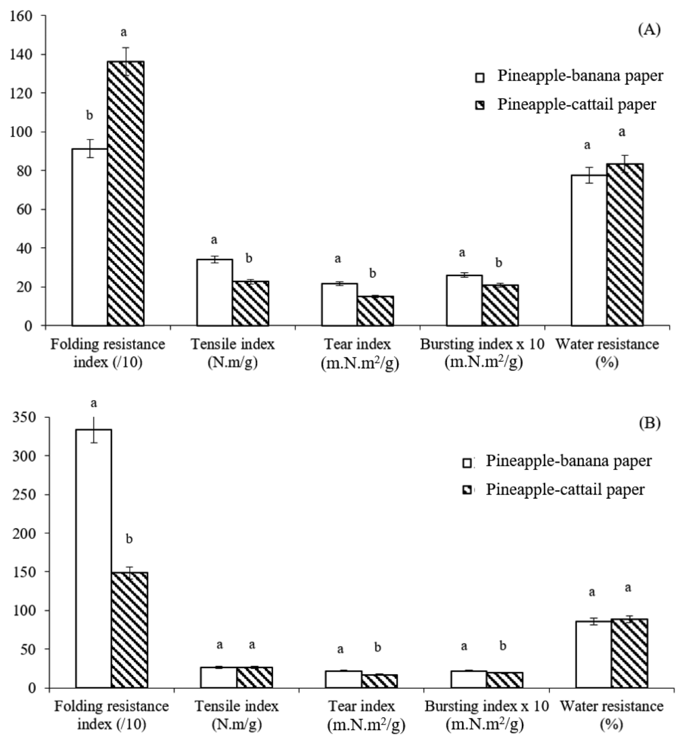

Fig. 4 Mechanical properties and water resistance of pineapple-banana and pineapple-cattail papers at 70:30 ratio coated with 0.6% chitosan and 2.0% water-resistant agent for: (A) 160 g/m^2; (B) 200 g/m^2, where different lowercase letters in same property indicate significant ($p < 0.05$) difference.

This study demonstrated the utilization of pineapple leaf, banana pseudostem and cattail stem as agricultural wastes along with weed pulp preparation for paper packaging. The results showed that the optimum conditions for the pulping and bleaching processes could be assessed based on characterization of the yield content and brightness of the pulp. Pulping with 30% NaOH followed by bleaching with 40% H_2O_2 was the best treatment for all the raw materials. Paper preparation at a ratio of 70:30, for either handmade papers produced from pineapple-banana pulps or pineapple-cattail pulps had appropriate mechanical properties, especially the tensile index. The pineapple-banana paper had better tensile and burst indices, as well as having water-resistant properties compared to the pineapple-cattail paper. The pineapple-banana papers had better mechanical properties when coated with 0.6% chitosan and 2.0% PFOA-free fluorocarbon as the optimum concentrations. Thus, the pineapple-banana mixed papers showed promise for use as paper packaging.

Conflicts of Interest

The authors declare that there are no conflicts of interest.

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