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Development and characterization of biocomposite films using banana pseudostem, cassava starch and poly(vinyl alcohol): A sustainable packaging alternative

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ABSTRACT

To meet the need for sustainable packaging, we introduce a novel biocomposite film consisting of banana pseudostem, cassava starch, and poly(vinyl alcohol). We aimed to evaluate the optimal biocomposite film composition, which is characteristic for packaging materials. Using the solvent casting method, we produced biocomposite films with varying proportions (10–40 % w/w) of the lignocellulosic component from both Sour and Ash Plantain banana pseudostems. The resulting biocomposite films were characterized for mechanical, chemical, thermal, water absorption, gas permeability, and morphological properties. At the 25 % lignocellulosic level, a notable drop ($P < 0.05$) in tensile strength and elongation was observed, while water absorption increased, and gas permeability decreased. Fourier Transform Infrared Spectroscopy analysis revealed insights into the structural attributes of lignocellulosic composites. Thermogravimetric analysis indicated an onset temperature of 120 °C for thermal degradation, confirming the biocomposite's thermal stability. A fundamental discovery emerged with the optimal composition at a 30 % pseudostem powder inclusion, offering an exceptional balance of tensile strength, elongation at break, water absorption, and gas permeability. This breakthrough holds significant implications for eco-friendly biocomposite films, particularly in food packaging. Future work may be undertaken to further explore banana pseudostems' potential in creating biocomposite films with advanced functionalities and their broader applications, including characterizations.

1. Introduction

Packaging materials have been essential in various industries, facilitating global product distribution. Plastics have seen increasing use, especially in the last few decades, due to industrialization and urbanization, resulting in a surge in single-use plastics for packaging (Singh et al., 2022). This extensive plastic usage has led to a significant issue, a massive 6.30 billion tons of plastic waste generated from 1950 to 2015, with just 9 % recycling and over 80 % ending up in landfills and ecosystems (Olivares Quispetera et al., 2021). The environmental impact of petroleum-based plastic waste is severe, primarily due to its non-biodegradable, non-recyclable nature, contributing to greenhouse gas emissions and climate change (Ford et al., 2022; Tian et al., 2020).

Conventional plastic packaging materials like polystyrene (PS), polyethylene terephthalate (PET), and polypropylene (PP), derived from non-renewable petroleum sources, resist natural degradation and pose health and environmental threats as they break down into microplastics (Buxoo & Jeetah, 2020). To address these issues, there is a growing interest in biodegradable and eco-friendly packaging materials. These materials, including starch-based polymers, polyhydroxyalkanoate (PHA), poly(lactic acid) (PLA), and chitosan-based films, offer biodegradability and good gas barrier properties, addressing the problems associated with traditional plastics (Jayarathna et al., 2022; Moshood et al., 2022; Perera et al., 2023).

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Starch, abundant and cost-effective, holds the potential for eco-friendly packaging (Falua et al., 2022). However, native starch's brittleness, high water sensitivity, and poor barrier properties limit its application (Amaraweera et al., 2021). Starch primarily consists of amylose and amylopectin (Bertoft, 2017), with cassava starch emerging as a staple food source. Despite its limited water resistance and mechanical properties compared to synthetic polymers, efforts have focused on blending starch with other polymers to overcome these limitations.

Poly(vinyl alcohol) (PVA) offers water solubility, non-toxicity, excellent gas barriers, and biodegradability, making it an attractive option. Blending PVA with starch aims to create biodegradable composite with improved mechanical and barrier properties. The study of the thermodynamic compatibility between starch and PVA is crucial, as partial miscibility can lead to phase separation and deteriorating mechanical properties (Abdullah & Dong, 2019; Iskalieva et al., 2023; Jayarathna et al., 2022; Patil et al., 2021). Molded Pulp Products (MPPs), primarily composed of cellulose and water, are eco-friendly packaging materials. They offer an alternative to fossil-based, non-biodegradable materials such as expanded polystyrene and vacuum-formed PET (Singh et al., 2022). However, MPPs using banana pseudostems as packaging material remain unexplored.

Banana (*Musa* spp.) cultivation generates a significant amount of banana pseudostem (BP) waste. This waste emits harmful gases, making field disposal unfeasible, and it lacks value as animal feed. The pseudostem consists of water, cellulose, hemicellulose, and lignin, making it suitable as a filler and reinforcement agent in composite materials (Hafizulhaq et al., 2018; Jayaprabha et al., 2011). This study explores the potential of a bioplastic composite comprising banana pseudostem, cassava starch, and PVA as an alternative to conventional plastic packaging. The objective is to assess the feasibility of producing PVA/cassava starch/banana pseudostem (BP) biocomposite films with varying BP compositions and their suitability for eco-friendly packaging. We hypothesized that incorporating different levels of BP into PVA/cassava starch biocomposite films can create biodegradable packaging with enhanced mechanical properties and compatibility between components, improving overall packaging quality.

2. Materials and methods

2.1. Banana pseudostems sample preparation

Banana pseudostems were sourced from five distinct cultivars: Sour Plantain (SP), Sugar Plantain, Ash Plantain (AP), Suwandel, and Rath Kesel, Gampaha, Sri Lanka, during the Maha season (September to March) of 2022. The pseudostems underwent rigorous processing, which involved cleaning, oven-drying at 60 °C for 72 h using a Yamato Scientific Co. Ltd oven, grinding into a 0.5 mm powder with a Retsch ZM 200 mill (Germany), and sieving with a manual strainer to achieve a particle size (diameter) of less than 180 μm.

2.2. Determination of banana pseudostem chemical composition

2.2.1. Proximate analysis

Proximate composition analyses were performed in adherence to the standards outlined in the Association of Official Analytical Chemists (AOAC) methods 920.36, 984.13, 948.22, 923.03 (2000), and AOAC (2006).

2.2.1.1. Dry matter and ash determination. The dry matter, ash, and moisture contents of the banana pseudostem powder were determined through a precise gravimetric method. Porcelain crucibles were preheated at 105 °C in the drying oven for one hour, then cooled in a desiccator, and their weights were recorded (W1 g). Approximately 1.0 g of banana pseudostem powder from each cultivar was placed in these crucibles, and their weights were recorded (W2 g). Subsequently, the samples were dried overnight at 105 °C, cooled in a desiccator, and their weights were recorded (W3 g). The dry matter percentage was calculated using the following formula:

$$\%\ \text{dry matter} = (\text{W3} - \text{W1})\text{g} * 100 / (\text{W2} - \text{W1})\text{g} \tag{1}$$

The dried samples were further utilized to determine the ash content. The crucibles with dried samples were placed in a Muffle Furnace (DMF-12, Human Lab Instrument Co., Korea) at 600 °C for 4 h. Following removal from the furnace and cooling in a desiccator, their weights were recorded (W4 g), and the ash percentage was calculated as follows:

$$\%\ \text{ash} = (\text{W4} - \text{W1})\text{g} * 100 / (\text{W3} - \text{W1})\text{g} \tag{2}$$

2.2.1.2. Moisture content. The moisture content of the banana pseudostem was computed using the equation:

$$\%\ \text{moisture} = [\text{Sample fresh weight(g)} - \text{Sample oven} - \text{dried weight(g)}] * 100 / \text{Sample fresh weight(g)} \tag{3}$$

2.2.1.3. Crude protein determination. The determination of crude protein contents in banana pseudostem powders was executed using the Micro Kjeldahl method. Approximately 0.25 g of banana pseudostem powder was digested in Kjeldahl tubes with 10 g of digestion mixture and 5 mL of concentrated sulphuric acid (H_2SO_4) for roughly an hour, or until the solution turned clear. After digestion, the tubes were allowed to cool to room temperature. Subsequently, a 4 % (v/v) boric acid solution was added to 250 mL conical flasks and placed on the distillation apparatus. Each Kjeldahl tube was also added with 5 mL of distilled water. Distillation continued until 100 mL of distillate was collected, which was then titrated against 0.1 N H_2SO_4. The crude protein content was calculated as follows:

$$\text{Crude Protein}(\%) = (\text{Burette reading(mL)} * \text{Normality of } H_2SO_4 * 8.75) / [\text{Weight of sample(g)} * \%\ \text{Dry Matter of the sample}] \tag{4}$$

2.2.1.4. Crude fat determination. The determination of the crude fat content of the banana pseudostem powder was carried out using the Soxhlet method (AOAC, 2000). Initially, clean and dry beakers were weighed (W1 g). Approximately 1 g of banana pseudostem powder was weighed and placed in a cellulose thimble. A volume of 310 mL of acetone was added to the fat-extracting beakers. The beakers were heated for 4–5 h to extract fat from the samples. After extraction, the beakers were weighed again (W_2 g). The crude fat percentage was calculated as follows:

$$\text{Crude fat}(\ \%) = [\text{Weight of beaker and fat (g)} - \text{Weight of empty dry beaker (g)}]*100 / [\text{Weight of dry sample (g)} - \%\text{Dry Matter of the sample}] \tag{5}$$

2.2.2. *Van Soest fiber components*

The Van Soest method (Van Soest & Knooren, 1997) was employed to determine the contents of Acid Detergent Fiber (ADF), Neutral Detergent Fiber (NDF), and Acid Detergent Lignin (ADL). These parameters were subsequently used to calculate hemicellulose and cellulose contents.

2.2.2.5. Acid detergent fiber (ADF). Approximately 1 g of the sample was placed in a Berzelius beaker, and 100 mL of an acid detergent solution, along with 2 mL of decalin, was added. The beakers were heated on a refluxing apparatus. Subsequently, crucibles were placed on a filtering apparatus, and the % ADF was calculated based on the following equations:

$$\text{Acid detergent fiber }\%\text{on a partial dry basis} = (\text{Weight of crucible and fiber } - \text{Weight of crucible})\text{g} * 100 / \text{Sample weight (g)} \tag{6}$$

$$\text{Adjusted to dry basis} = \text{Acid detergent fiber }\%\text{ of partial dry sample} * 100/ \text{ Dry matter }\%\text{ of partial dry sample} \tag{7}$$

2.2.2.6. Neutral detergent fiber (NDF). Approximately 1 g of the sample was placed in a Berzelius beaker, and 100 mL of a neutral detergent solution, along with 2 mL of decalin, was added. The beakers were heated on a refluxing apparatus, with sodium sulfite added. After boiling for 5–10 min, crucibles were placed on a filtering apparatus. The % NDF was calculated based on the following equations:

$$\text{Cell wall }\%\text{ on a partial dry basis} = (\text{Weight of crucible and cell walls } - \text{Weiht of crucible})\text{g} * 100 / \ \text{Sample weight (g)} \tag{8}$$

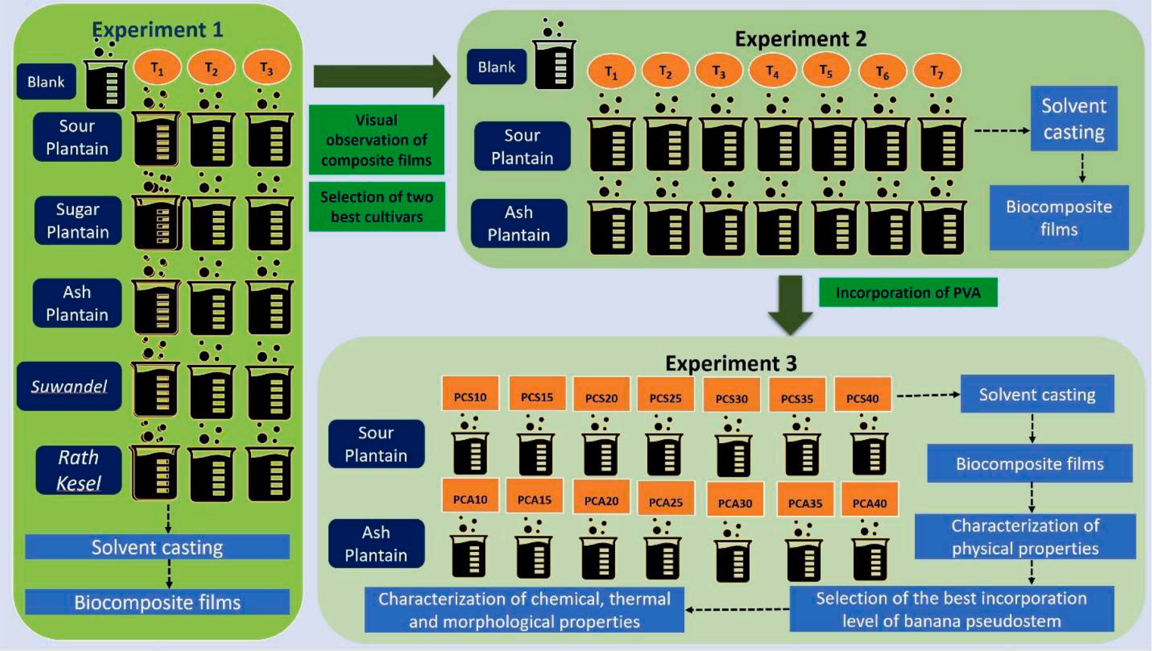

Fig. 1. A schematic outline of the study representing Experiments 1, 2 and 3. T_1-T_7 are different treatments applied in experiment 1 and 2. PCS10-PCS40 and PCA10 PCA40 represent the different treatments of Sour Plantain and Ash Plantain respectively in Experiment 3, where abbreviations for the composite samples are given based on the following strategy; P = PVA, C = Cassava starch, S = Sour Plantain, A = Ash Plantain and numbers indicates the poly(vinyl alcohol) content (%).

Table 1
Composition of bioplastic samples for 3 treatments.

Sample	Cassava starch (g)	Lignocellulose of banana pseudostem (g)	% Mass of lignocellulose material	% Mass of starch	Lignocellulose/starch ratio
Blank	7	0.0	0.0	100.0	0/7
Treatment 1	7	1.0	12.5	87.5	1/7
Treatment 2	7	1.5	17.7	82.4	1.5/7
Treatment 3	7	2	22.2	77.8	2/7

Table 2
Compositions of bioplastic samples of 7 treatments.

Sample	Cassava starch (g)	Lignocellulose content (g)
Treatment 1	7.0	0.2
Treatment 2	7.0	0.4
Treatment 3	7.0	0.6
Treatment 4	7.0	0.8
Treatment 5	7.0	1.0
Treatment 6	7.0	1.2
Treatment 7	7.0	1.4
Blank	7.0	-

$$\text{Adjusted to dry basis} = \text{Cell wall \% of partial dry sample} * 100/\ \text{Dry matter \% of partial dry sample} \quad (9)$$

2.2.2.7. Acid detergent lignin (ADL). ADL content of banana pseudostems was determined using the samples obtained in the final step of determining the ADF content. The crucibles were filled with cooled 72 % sulphuric acid and stirred. After multiple refills, the acid was filtered, and the residues were rinsed with hot water. The crucibles were dried, ignited, and ADL content was calculated based on the following equations:

$$\text{Lignin (\%) on a partial dry basis} = (\text{Crucible and lignin weight} - \text{Crucible and ash weight})\text{g} * 100/\ \text{Sample weight (g)} \quad (10)$$

$$\text{Adjusted to dry basis} = \text{Lignin \%on a partial dry sample} * 100/\text{Dry matter \% of a partial dry sample} \quad (11)$$

Table 3
Composition of biocomposite films.

Sample	PVA (%)	Pseudostem Powder (%)	Cassava starch (%)	Glycerol (as a % of dry cassava starch)
PCS10	10	10	5	30
PCS15	15	15	5	30
PCS20	20	20	5	30
PCS25	25	25	5	30
PCS30	30	30	5	30
PCS35	35	35	5	30
PCS40	40	40	5	30
PCA10	10	10	5	30
PCA15	15	15	5	30
PCA20	20	20	5	30
PCA25	25	25	5	30
PCA30	30	30	5	30
PCA35	35	35	5	30
PCA40	40	40	5	30

Abbreviations for the composite samples are given based on the following strategy; where P = PVA, C = Cassava starch, S = Sour Plantain, A = Ash Plantain

This comprehensive analysis procedure enabled the precise determination of the chemical composition of banana pseudostems, contributing valuable insights into their potential use in preparations of bioplastic composites as described below.

2.3. Preparation of the bioplastic composite

A general outline of the study design is presented in Fig. 1.

Experiment 1. Preparation of Cassava starch/banana pseudostem biocomposite film using 3 treatments

Bioplastic composites were created using a solution casting method, following the approach by Quispetera et al. (2021) with slight modifications s described below. Industrial-grade cassava starch was used, with glycerol as the plasticizer and 3–5 % acetic acid. Approximately 7 g of cassava starch was mixed with 1 mL of acetic acid, 1 mL glycerol, and 50 mL of distilled water. Different doses (1.0 g, 1.5 g, 2.0 g) of banana pseudostem powder were incorporated as lignocellulosic fillers and homogenized at 70 °C. This formulation process was applied to all five banana cultivars. The resulting bioplastic material was cast into 8 cm diameter Petri dishes and allowed to evaporate naturally at temperatures of 25–27 °C for 24 h. Subsequently, the samples were oven-dried at 60 °C for 6 h. After drying, the biocomposite films were carefully detached from the Petri dishes. Additionally, a cassava starch bioplastic film (Blank) was fabricated and used as a reference point (Table 1).

Experiment 2. Preparation of cassava starch/banana pseudostem biocomposite films using 7 treatments

The same method as employed in Experiment 1 was replicated with the modification of seven different treatments to select the most appropriate composition to develop the bioplastic composites using the pseudostem powder of Sour plantain (SP) and Ash Plantain (AP), the selected varieties from Experiment 1. Each of the treatments was incorporated with 7 g of cassava starch, 1 mL of acetic acid, 1 mL glycerol and 50 mL of distilled water and banana pseudostem powder as the lignocellulose content in a sequence of 0.2 g, 0.4 g, 0.6 g, 0.8 g, 1.0 g, 1.2 g and 1.4 g respectively. The specific compositions for each treatment are mentioned in Table 2.

Experiment 3. Preparation of poly(vinyl alcohol) (PVA)/Cassava Starch/Banana Pseudostem Biocomposite films

Poly(vinyl alcohol) with a polymerization degree of 1700 and a hydrolysis degree of 99 %, as well as glycerol, were procured from Sigma Aldrich Co., St. Louis, MO, USA. A predetermined quantity of PVA was mixed with distilled water to create a solution with a solid content of 10 %. This solution was heated to 90 °C while continuously stirring until a homogeneous solution was achieved. Distilled water was added as necessary to compensate for any moisture loss during heating. The biocomposite film were prepared using the solution casting method described by Imam et al. (2005) with slight modifications. The composite material consisted of 5 % cassava starch (w/w), 30 % glycerol (w/w, relative to dry starch), and an appropriate amount of banana pseudostem powder (either Sour Plantain pseudostem powder or Ash Plantain pseudostem powder) along with PVA (tenfold the quantity of banana pseudostem powder) as described in Table 3. Unlike in Experiment 1 and 2, acetic acid was not incorporated in Experiment 3, and PVA was used as the compatibilizing agent of cassava starch. The main purpose of Experiment 3 was to find the effect of PVA in the film-forming mixture. The number of treatments used were as similar as in Experiment 2. PVA, cassava starch, and glycerol were mixed and the resulting mixture was heated at 80 °C for 30 min under continuous stirring. Following this, banana pseudostem powder was added, and the mixture was stirred for an additional 10 min. Once the solution was homogenized, the heat source was turned off, and approximately 25 g of the aqueous solution were cast onto Petri dishes. The samples were left to undergo solvent evaporation overnight at ambient temperature (25–27 °C). Subsequently, the samples were placed in a drying oven at 60 °C for 6 h. Following the drying process, the samples were allowed to cool to room temperature, and the biocomposite films were carefully peeled from the Petri dishes. The average thickness of the samples was approximately 0.30 mm. The compositions of the biocomposite films are detailed in Table 3.

2.4. Characterisation of biocomposite films

2.4.1. Mechanical properties: tensile strength and elongation at break

Tensile strength of the prepared biocomposite film was determined following the standard procedure outlined in ASTM, International, 2009, with slight adjustments as detailed by Amaraweera et al. (2021). The tensile strength test was conducted employing the INSTRON 3365 Universal Testing Machine (Instron Ltd, Buckinghamshire, UK) with a fixed crosshead speed of 10 mm/min. Samples were crafted in accordance with the dimensions stipulated by ASTM, International, (2009). Three specimens were cut from each film, with a sample length and width of 12 cm and 6 cm, respectively. The sample thickness was maintained at 0.25 mm. During the stretching of these specimens, measurements for tensile strength (in MPa) and elongation at break (in %) were recorded.

2.4.2. Chemical properties

The functional groups and chemical bonds of the biocomposite film samples were characterised using Fourier Transform Infrared Spectroscopy (FTIR) following the approach described by Amaraweera et al. (2021) with minor modifications. Characteristic peaks for Sour Plantain, Ash Plantain, cassava starch, and PVA/cassava starch/lignocellulosic biocomposite film were obtained using an FTIR spectrometer (PerkinElmer, USA) equipped with an ATR reflectance cell. Spectra were collected within the wavenumber range of 500 to 4000 cm^{-1}. Two scans were obtained at a resolution of 4 cm^{-1} for each sample. FTIR spectra were analysed using MS Excel 2019 software.

2.4.3. Thermal properties

Thermal degradation and moisture content of the biocomposite film samples were evaluated using Thermogravimetric Analysis (TGA). A TGA 5500 (TA Instruments, USA) was employed for the analysis under atmospheric air. Platinum crucibles of 100 μL were utilised with a heating rate of 10 °C/min, maintaining the temperature within the range of 25 °C to 700 °C. The TRIOS software (version 4.4.0, TA Instruments, USA) was used to calculate the thermal degradation temperature and moisture content of the biocomposite films. The TGA spectra were analysed using the OriginPro 2018 software.

Table 4
Results of proximate and ADF/ NDF/ADL constituents (DM basis) of the banana pseudostem powders.

Constituent	Sour plantain	Sugar Plantain	Ash plantain	*Suwandel*	*Rath Kesel*
% DM	92.7 ± 0.1^{a}	90.8 ± 0.0^{a}	92.2 ± 0.0^{a}	93.2 ± 0.2^{a}	90.4 ± 0.1^{a}
% Ash	8.0 ± 0.7^{a}	10.8 ± 0.7^{b}	13.3 ± 0.1^{c}	10.3 ± 0.4^{a}	9.2 ± 0.1^{a}
% Moisture	7.3 ± 0.2^{a}	9.2 ± 0.5^{b}	7.8 ± 0.3^{a}	6.8 ± 0.4^{a}	9.6 ± 0.6^{b}
% CP	2.8 ± 0.2^{a}	3.0 ± 0.0^{b}	2.3 ± 0.6^{a}	2.4 ± 0.3^{a}	3.7 ± 0.3^{c}
% CF	5.1 ± 0.9^{b}	5.1 ± 0.6^{b}	4.8 ± 0.8^{a}	5.6 ± 0.4^{c}	4.1 ± 0.6^{a}
% ADF	38.5 ± 0.4^{a}	37.9 ± 0.2^{a}	37.6 ± 0.3^{a}	41.5 ± 0.9^{b}	45.7 ± 0.9^{c}
% NDF	63.5 ± 0.7^{b}	55.9 ± 0.7^{a}	59.3 ± 0.3^{a}	67.8 ± 0.3^{c}	66.1 ± 0.5^{c}
% ADL	36.7 ± 0.3^{ab}	33.9 ± 0.4^{a}	39.9 ± 0.7^{b}	35.8 ± 0.3^{a}	41.0 ± 0.6^{c}

Values are mean ± SD based on the average of triplicates. Different superscript letters (a, b and c) in the same row indicate a significant difference ($P < 0.05$) among treatments.

2.4.4. Water absorption

The water absorption properties of banana pseudostem-incorporated biocomposite films were measured following the standard method described by ASTM International (2010). The samples were conditioned at 50 °C for 24 h and weighed (M_o) before testing. Subsequently, the samples were immersed in distilled water. After 2 h, the samples were removed from the water, dried with a clean, dry cloth, and weighed immediately (M1). Then, the samples were soaked again in water for another 22 h and weighed (M2). The water absorption capabilities of the thin films were calculated using the following equations:

$$\text{Water absorption (\%)at 2 hours} = (M1 - M_o)g * 100 / Mog \tag{12}$$

$$\text{Water absorption (\%) at 24 hours} = (M2 - M_o)g * 100 / Mog \tag{13}$$

2.4.5. Gas permeability

Gas permeability was assessed following the standard procedure described by ASTM 1434-82. Gas permeability of the biocomposite film was determined using the air permeability tester (Labthink Instruments Co., Ltd., China) with a constant pressure difference between the two sides of the specimen. The air permeability was measured by calculating the air flux through a specific area within a certain time frame. The temperature and humidity were maintained at 23 °C and RH 80 %, respectively.

2.4.6. Morphological properties

The morphology of the banana pseudostem-incorporated biocomposite film was visualised using Scanning Electron Microscopy (SEM) as outlined by Amaraweera et al. (2021). The samples were sputter-coated with gold for 1 min before imaging and examined at an acceleration potential of 5 kV using SEM (ZEISS, Germany). The morphological investigations were conducted under x 1000 magnification.

2.5. Statistical analysis of data

The experimental design employed for the statistical analysis of data was the Completely Randomized Design (CRD). All parametric data (except for the SEM) were analysed using SAS 9.0 version. The results were expressed as the mean ± standard error of the mean. One-way analysis of variance (ANOVA) and Duncan's Multiple Range Test were utilised to determine the significant differences among different treatments. Differences were considered significant at $P < 0.05$

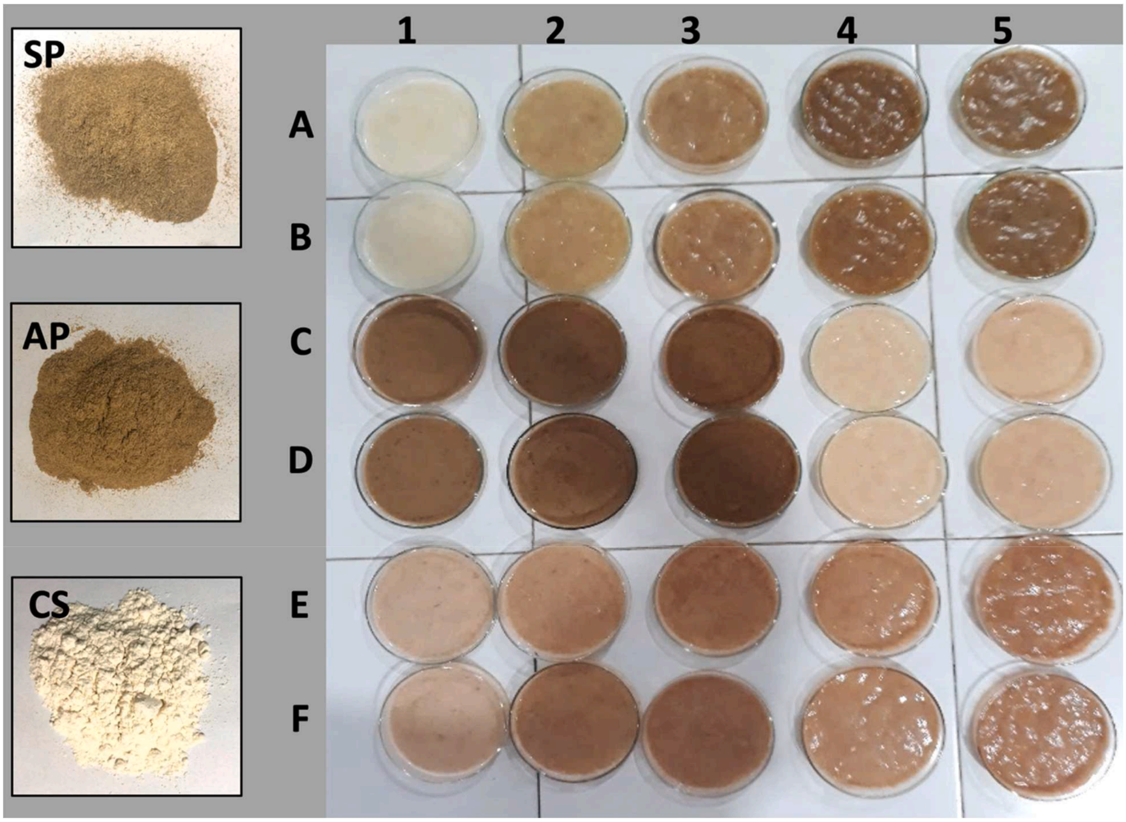

Fig. 2. Preparation of cassava starch/banana pseudostem biocomposite film and neat cassava films. Combinations are: A1/B1-Neat Cassava, A2/B2-0.2 g SP, A3/B3-0.4 g SP, A4/B4-0.6 g SP, A5/B5-0.8 g SP, C1/D1-1.0 g SP, C2/D2-1.2 g SP, C3/D3-1.4 g SP, C4/D4-0.2 g AP, C5/D5-0.4 g AP, E1/F1-0.6 g AP, E2/F2-0.8 g AP, E3/F3-1.0 g AP, E4/F4-1.2 g AP, E5/F5-1.4 g AP Key ingredients of bioplastic composite material used in the experiment; SP: Sour Plantain pseudostem powder, AP: Ash Plantain pseudostem powder, CS: Cassava starch

3. Results and discussion

3.1. Chemical composition of banana pseudostems

Analyzing the constituents of banana pseudostems is essential for understanding the properties of biocomposite films. For this purpose, we conducted proximate analysis and Van Soest constituent tests on BP powders from five selected banana cultivars. The results of the dry matter (% DM), ash (% Ash), crude protein (% CP), crude fat (% CF), acid detergent fiber (% ADF), neutral detergent fiber (% NDF), and acid detergent lignin (% ADL) tests for these banana cultivars are presented in Table 4.

The ADF content accounts for the cellulose and lignin content of banana pseudostems. The NDF content comprises cellulose, hemicellulose, and lignin components (Salem, 2019). The difference between the NDF and ADF values of a sample provides the hemicellulose content.

All the selected cultivars showed no significant difference in DM content. The highest percentage of ash was reported for Ash Plantain, indicating a distinct composition. Generally, banana pseudostems are characterized by a high content of lignocellulosic compounds, lower CP (%) and CF (%) compared to mature banana leaves. According to Ingale et al. (2014), the chemical composition of banana pseudostems includes 30.3-35.2 % cellulose, 19.3–24.4 % hemicellulose, 2.5–3.4 % lignin, and 7.3–10.7 % ash content. In another study, Srivastava et al. (2019) reported that BP fiber consists of 65 % cellulose, 6 % hemicellulose, 13 % lignin, and 14 % ash. However, the current study, for all selected cultivars, reported average amounts of 32.5 %, 23.5 %, and 3.1 % for cellulose, hemicellulose, and lignin contents, respectively. Incorporating lignocellulosic material or fibre is essential as a filler or reinforcement agent in starch-based biocomposite film to enhance their mechanical properties.

Cellulose, hemicellulose, and lignin are the major constituents accounting for plant fibre composition (Srivastava et al., 2019). Lignin is the most abundant organic compound in plants, second only to cellulose (Moura et al., 2010). The selected banana pseudostem samples exhibited varying compositions in terms of % ADF, % NDF, and % lignin, with *Rath Kesel* reporting the highest ADF and ADL values.

3.2. Preparation of cassava starch/banana pseudostem biocomposite films (3 Treatments)

Chemical treatments using acid or alkali can be applied to modify the fibre matrix adhesion, resulting in better tensile strength in the biocomposite film (Srivastava et al., 2019). In this study, BP powders were pretreated with acetic acid and incorporated into the mixtures. Glycerol served as the plasticizer, while cassava starch and pseudostem powder acted as the binder and filler/reinforcement agent, respectively. The resulting samples from the solvent casting method were compared for their uniformity in appearance by visual observation in order to determine the most appropriate two cultivars of banana to continue with the study. As a result, Sour Plantain and Ash Plantain were selected as the better options among the cultivars in terms of appearance and therefore, opted to be further tested in this study. Furthermore, treatments with 1.5 g of BP powder incorporation resulted in improved uniformity in appearance in biocomposite films.

3.3. Preparation of cassava starch/banana pseudostem biocomposite films (7 Treatments)

Following the results from the previous study, we prepared a series of pseudostem compositions comprising seven treatments (0.2 g, 0.4 g, 0.6 g, 0.8 g, 1.0 g, 1.2 g, 1.4 g) in duplicate from both Sour Plantain (SP) and

Fig. 3. Morphological illustrations of the biocomposite film films composed of the following components: 30 % PVA, 30 % Sour Plantain Pseudostem Powder, 5 % cassava, and 30 % glycerol based on dry cassava starch percentage, as detailed in Table 3 and referred to as PCS30 biocomposite film.

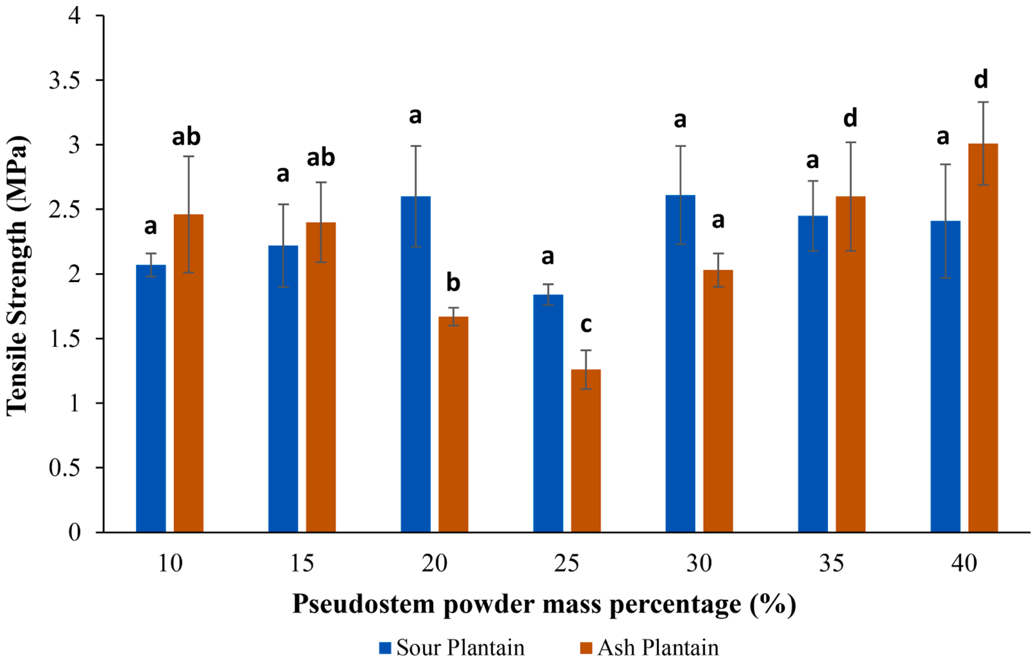

Fig. 4. Tensile strength of biocomposite film with different pseudostem powder mass percentages in composites formula. Values are mean ± SD based on the average of triplicates. $^{a\text{-}d}$ Means with different letters on the top of bars are significantly ($P < 0.05$) different among treatments.

Ash Plantain (AP) pseudostem powders to determine the most appropriate composition for developing bioplastic composites (Fig. 2). During these experiments, certain samples were subjected to mould growth during the 24 h air-drying process, possibly due to the high moisture content retained in the biocomposite film. In some cases, it was challenging to peel the biocomposite films off from the glass Petri dishes, which could be attributed to over-drying or the use of high temperatures over a short period during drying, leading to rapid moisture evaporation and surface cracks. Moreover, biocomposite film tended to shrink and fold upon themselves due to the higher drying rate on the exposed surface compared to the surface in contact with the casting plates in agreement with Srivastava et al. (2019).

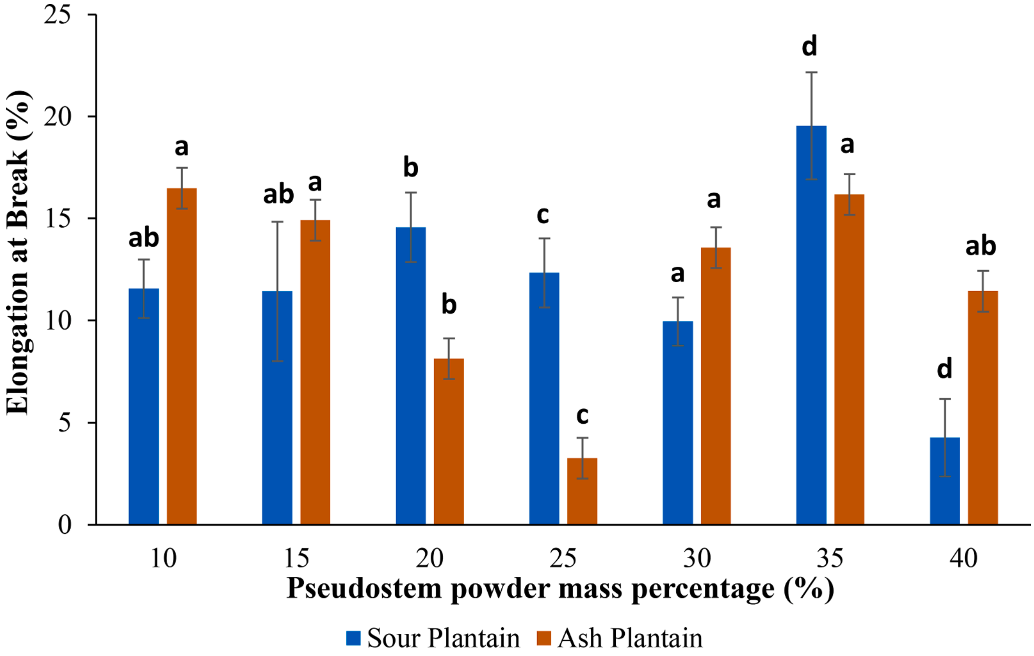

Fig. 5. Percentage elongation of biocomposite film with different pseudostem powder mass percentages in composites formula Values are mean ± SD based on the average of triplicates. $^{a–d}$ Means with different letters on the top of bars are significantly ($P < 0.05$) different among treatments.

3.4. Preparation of poly(vinyl alcohol) (PVA)/cassava starch/banana pseudostem biocomposite films

The same pseudostem compositions used in Experiment 2 were employed with different ingredients. We incorporated the biodegradable polymer into the PVA into the mixtures as a compatibilizing agent for cassava starch. Starch primarily contributes to the water absorption of the blends due to its inherent hydrophilic nature (Jayarathna et al., 2022). Increasing PVA content reduced the water absorption capacity of the biocomposite film, as reported by Judawisastra et al. (2017). They found that neat tapioca starch had a maximum water absorption of 495 %, while PVA had a maximum of 43 %. Blending cassava starch with a synthetic biodegradable polymer offers cost reduction, improved biodegradation rate, compatibility, renewability, and synergetic effects (Gunawardene et al., 2021).

The addition of PVA to the biocomposite film material improved uniformity in the texture compared to Experiments 1 and 2. Furthermore, PVA-incorporated biocomposite film were easier to peel off from the glass trays or Petri dishes compared to films without PVA. Fig. 3 presents biocomposite film made with 30 % Sour Plantain pseudostem incorporation (PCS30). We maintained a constant volume of the film-forming solution to ensure a film thickness of 3 mm. The surface smoothness of the film depended on the particle size of BP used, as observed in SEM image analysis in a previous study (Rizal et al., 2022). In another study, glycerol as the plasticizing agent imparted elasticity to the biocomposite film, making them more flexible with an increased

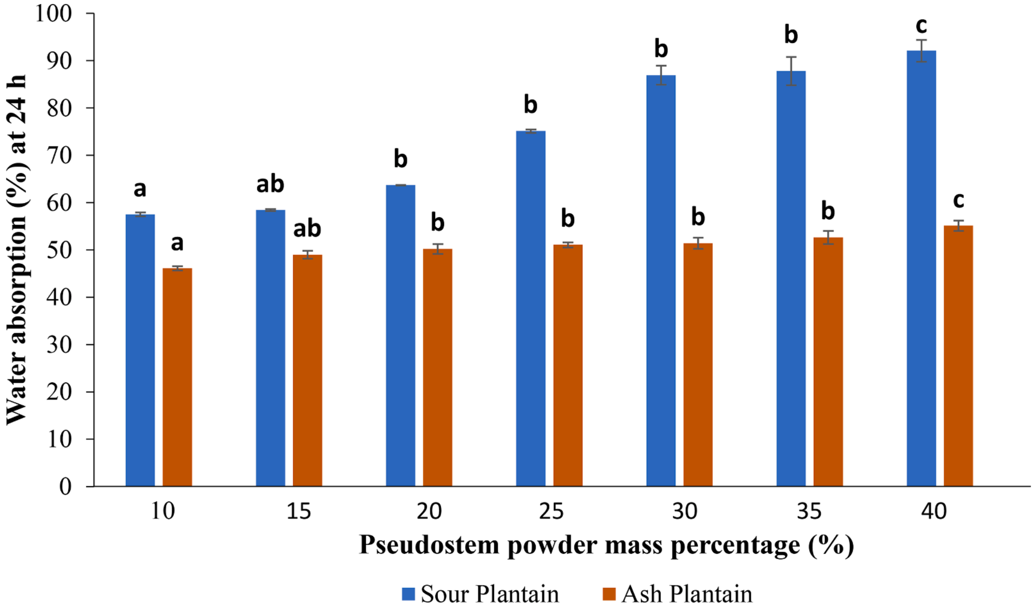

Fig. 6. Water absorption (%) of biocomposite films within a duration of 24 h. Values are mean ± SD based on the average of triplicates. $^{a–c}$ Means with different letters on the top of bars are significantly ($P < 0.05$) different among treatments.

percentage elongation at break (Nasir & Othman, 2021).

3.5. *Mechanical properties of biocomposite films*

3.5.1. *Tensile strength*

Fig. 4 presents the alterations in the tensile strength of biocomposite films. The tensile strength of biocomposite film, incorporating various proportions of Sour Plantain BP, showed no significant differences. However, the lowest ($P < 0.05$) tensile strength value, at 1.3 ± 0.2 MPa, was observed when 25 % lignocellulose from Ash Plantain was added. This may be attributed to non-uniformity in the film-forming mixture resulting from the diverse particle sizes of banana pseudostem powder, as well as the accumulation of a greater mass of oxygen molecules within the bioplastic films (Asyraf et al., 2021). Conversely, the highest tensile strength value ($P < 0.05$) was achieved with a 40 % incorporation of Ash Plantain BP.

Quispetera et al. (2021) reported that the inclusion of a lignocellulose mass percentage of 22.22 % increased the tensile strength of the bioplastic film made from *Manihot esculenta* and *Ficus benjamina* by up to 58.59 %, compared to the neat cassava starch bioplastic film. In a similar study, Jaafar (2018) noted a 42 % increase in tensile strength with the addition of 30 % pineapple leaf fibre in the production of tapioca bioplastic.

Gonzalez et al. (2018) reported improved tensile strength when incorporating 15 % wood sawdust in a green composite packaging film utilising BP and PVA. Furthermore, Espina, Cruz-Tirado, and Siche (2016) determined that an optimal concentration of agro-industrial fibres is 15 % in starch-based composite formulations. Several studies have indicated a decline in tensile properties in bioplastic films as the level of lignocellulose filler surpasses a certain threshold (Espina et al. 2016; Olivares Quispetera et al., 2021). Hence, it is imperative to establish the optimum lignocellulose proportion that yields superior mechanical properties.

In the current study, the 25 % incorporation level displays a sudden drop in the tensile strength which is supposed to increase along with the increase of BP incorporation level. Adhering to the study results of Jaafar (2018) and considering the tensile strength results of this study, 30 % BP incorporation level was finalized ideal.

3.5.2. *Elongation at break*

The maximum percentage of elongation (as shown in Fig. 5) was achieved when 30 % of Sour Plantain BP was added to the biocomposite film, while the lowest ($P < 0.05$) percentage of elongation was observed with a 25 % incorporation level of Ash Plantain (AP). In a similar study, an increase in lignocellulose content led to a reduction in percentage elongation in biocomposite film (Olivares Quispetera et al., 2021). Another study noted that the addition of cellulose and carboxymethylcellulose to starch-based bioplastics decreased the elongation at break from 42.2 % to 25.8 % (Abe et al., 2021). Considering the results of the analysis, it was concluded that the 30 % incorporation level with the highest percentage of elongation to be the ideal.

3.5.3. *Water absorption*

The water absorption (%) of biocomposite film over a 24 h period is depicted in Fig. 6. The water absorption (%) at 24 h for the biocomposite film increased as the lignocellulose mass percentage increased. Specifically, the biocomposite film with a 40 % Sour Plantain lignocellulose mass percentage exhibited the highest water absorption ($P < 0.05$), reaching 92.1 ± 2.3 %. Although all the bioplastic softened after water absorption, they retained their integrity, maintaining most of their cohesiveness and consistency. In a previous study, it was reported that water absorption reached 74 % after 144 h of immersion, compared to 58.55 % in films without lignocellulose incorporation (Mittal et al., 2020).

Attributing a high potential for water absorption is not desirable with the packaging materials. In the study, the water absorption (%) increased with the increase of BP mass percentage. Therefore, the incorporation levels with the highest water absorption (%) have to be neglected at this point and 30 % incorporation level was selected ideal.

3.5.4. *Gas permeability*

Gas impermeability is a critical property for food packaging films to protect packaged food items from deterioration due to oxygen and water vapour exposure. In this study, gas permeability decreased as the lignocellulose content increased (Fig. 7). Notably, both Sour Plantain and Ash Plantain demonstrated similar gas permeability values at their respective lignocellulose mass percentages. This phenomenon can be attributed to lignin's role as a chemical adhesive among other fibres. Additionally, the coexistence of fibres with varying diameters can create intricate pathways for gas molecules (Brody et al., 2008).

In conclusion, the BP incorporation levels with high and too low gas permeability values were neglected and the 30 % incorporation level of

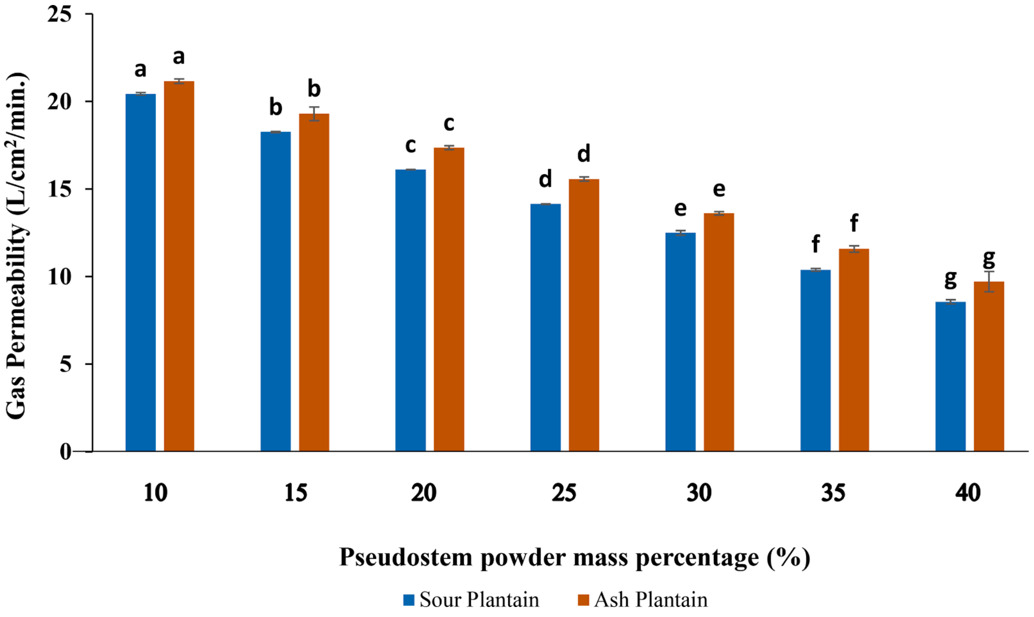

Fig. 7. Gas permeability of biocomposite films with pseudostem powder mass percentage.

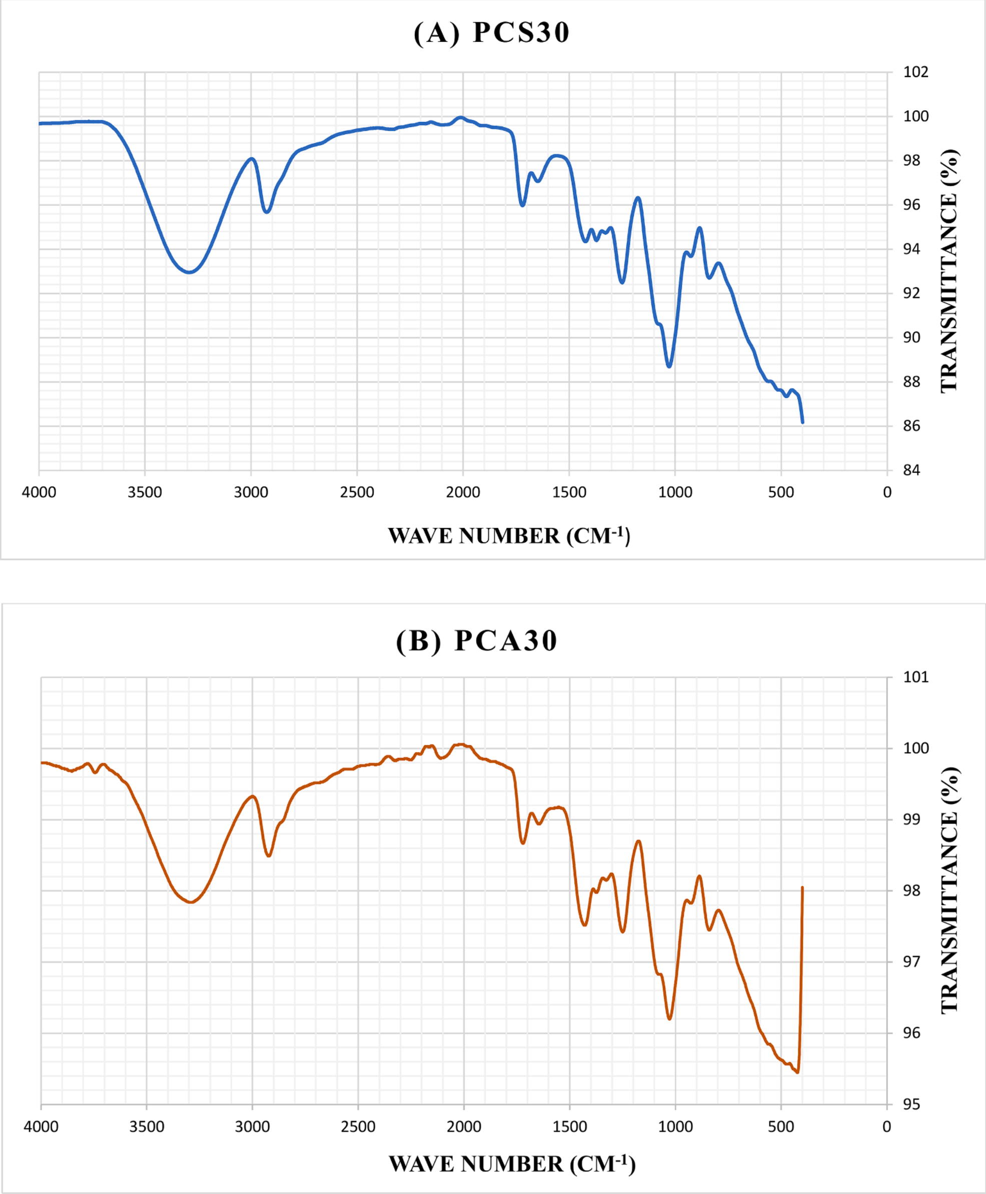

Fig. 8. FTIR spectra for the 30 % banana pseudostem (plot A: sour plantain and plot B: ash plantain) incorporated biocomposite films, where P = PVA, C = Cassava starch, S = Sour Plantain, A = Ash Plantain as detailed in Table 3.

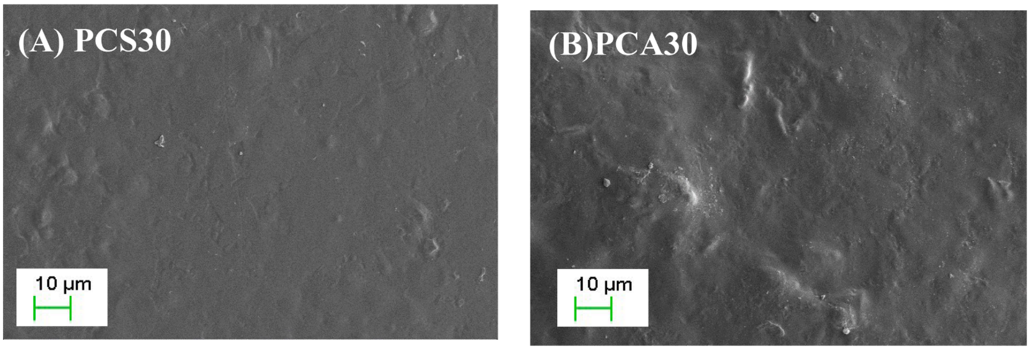

Fig. 9. SEM micrographs of PCS30 and PCA30 biocomposite films at X1000 magnification. Micrograph A: sour plantain and micrograph B: ash plantain incorporated biocomposite film, where P = PVA, C = Cassava starch, S = Sour Plantain, A = Ash Plantain as detailed in Table 3.

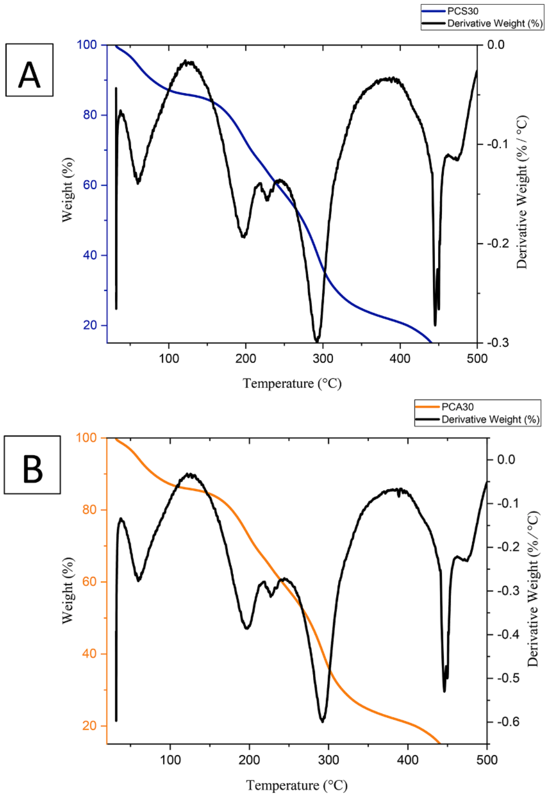

Fig. 10. Thermogravimetric analysis for PCS30 (A) and PCA30 (B) biocomposite film sample, where P = PVA, C = Cassava starch, S = Sour Plantain, A = Ash Plantain as detailed in Table 3.

BP was opted ideal in terms of gas permeability attributes of the biocomposite films.

Values are mean ± SD based on the average of triplicates. $^{a–g}$ Means with different letters on the top of bars are significantly ($P < 0.05$) different among treatments.

By considering the combined findings related to tensile strength, percentage elongation, 24 h water absorption, and gas permeability and the findings of previous research, it was determined that treatments with 30 % lignocellulose incorporation from both Sour Plantain and Ash Plantain (PCS30 and PCA30) are suitable for producing the recommended biocomposite films material for packaging. Subsequent analyses were conducted to assess the chemical, thermal, and morphological properties of the PVA/cassava starch-incorporated biocomposite film with 30 % BP lignocellulose incorporation.

3.6. *Characterization of chemical properties of biocomposite films*

3.6.1. *Fourier transform infrared spectroscopy (FTIR)*

The FTIR spectra of PCS30 and PCA30 as depicted in Fig. 8 below, provide insights into the functional groups and chemical bonds present in the biocomposite film with 30 % BP incorporation. In Fig. 8(A), two major peaks are observed at wave numbers of 1587 and 2000 cm^{-1}. The peak at 1587 cm^{-1} is attributed to the stretching of C=O bonds in ketone or carbonyl groups found in hemicellulose and lignin. Additionally, the peak at 2000 cm^{-1} is associated with the vibrational stretching of C-H bonds in methyl and methylene groups present in cellulose and hemicellulose.

In Fig. 8(B), two major peaks are observed at wave numbers of 1539 and 2000 cm^{-1}. The peak at 1539 cm^{-1} is attributed to the stretching of C=O bonds in ketone or carbonyl groups found in hemicellulose and lignin. The peak at 2000 cm^{-1} could be attributed to the vibrational stretching of C-H bonds in methyl and methylene groups present in cellulose and hemicellulose.

3.7. *Morphological properties: scanning electron microscopy (SEM)*

The SEM micrographs at X1000 magnification reveal that both PCS30 and PCA30 biocomposite film exhibit relatively uniform surface morphologies (Fig. 9). However, the morphology of PCS30 appears to be homogenous and continuous without any granules or pores, which is favourable in terms of packaging material attributes. The variations observed on the surface of the PCA30 with discontinuous and porous structure could be attributed to the uneven mixing of the film-forming solution (Srivastava et al., 2019).

3.8. *Thermal properties: thermogravimetric analysis (TGA)*

Thermogravimetric (TG) and its derivative thermogravimetric (DTG) curves were used to evaluate the thermal stability of the PCS30 and PCA30 biocomposite films. According to Fig. 10(A) below, the presence of multiple decomposition peaks aligns with the composite nature of the materials. When the temperature was below 100 °C, a lower thermal weightlessness was observed. This could be attributed to the evaporation of water or the small molecular impurities in the biocomposite films. Following an initial water loss of approximately 10 %, three degradation peaks were observed at 125 °C, 225 °C, and 388 °C in the PCS30 sample. The onset temperature (T_o) for the PCS30 sample was determined to be 120°C. Fig. 10(B), displays the thermogravimetric behavior of PCA30 biocomposite film. Following an initial approximate water loss of 10 %, three degradation peaks were observed at 125 °C, 218 °C, and 386 °C. The onset temperature (T_o) for the PCA30 sample was also determined to be 120 °C.

According to the graphs, the decrease in thermal stability is likely attributed to the initial decomposition of low molecular weight components, such as glycerol incorporated in the composite. In a similar study, Imam et al. (2005) reported three degradation peaks occurring after a water loss of around 4 %, at 221 °C, 320 °C, and 408 °C, with a T_o observed at 168 °C.

4. Conclusion

In conclusion, this research underlines the promising utilization of banana pseudostems in the development of biocomposite films, showcasing their rich lignocellulosic composition and potential in creating biocomposite film materials to yield diverse mechanical and physicochemical properties. The optimal composition was identified at a 30 % pseudostem powder inclusion level, resulting in exceptional mechanical and physicochemical properties suitable for eco-friendly bioplastic films, particularly in food packaging. Fourier Transform Infrared Spectroscopy analysis confirmed the successful integration of banana pseudostem into the biocomposite films, while morphological analysis indicated superior film formation in Sour Plantain biocomposite films. Thermal stability analysis revealed multiple degradation peaks with an onset temperature of approximately 120 °C, primarily attributed to low molecular weight constituents. These findings open avenues for sustainable biocomposite film materials, offering customizable properties for various industries, notably in biodegradable packaging. Further research and development are crucial to fully exploit the potential of banana pseudostems in biocomposite film material production and industrial applications.

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During the preparation of this work, the authors used ChatGPT to improve the language and readability with caution. After using this tool, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

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H.A.S. Dilkushi: Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Shishanthi Jayarathna:** Writing – review & editing, Writing – original draft, Validation, Supervision, Software, Resources, Project administration, Methodology, Data curation, Conceptualization. **A. Manipura:** Writing – review & editing, Validation, Supervision, Software, Resources, Investigation, Formal analysis. **H.K.B.S. Chamara:** Validation, Supervision, Resources, Investigation, Formal analysis, Data curation. **D. Edirisinghe:** Validation, Software, Resources, Methodology, Investigation, Formal analysis, Data curation. **J.K. Vidanarachchi:** Writing – review & editing, Writing – original draft, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Conceptualization. **Hasitha Priyashantha:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Software, Resources, Project administration, Methodology, Data curation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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