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Development of edible food coating using cassava starch/agar blends infused with clove oil for the preservation of mangoes

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Food coatings incorporating natural compounds are increasingly recognized for their effectiveness in preserving fresh produce from post-harvest decay and extending its shelf life. In this study, an edible bioplastic food coating was developed using cassava starch, agar and clove oil (CO) for the preservation of tropical fruits, specifically mango. The physicochemical and mechanical properties, water vapor transmission rate (WVTR), moisture absorption, solubility, biodegradability, carbon dioxide evolution, antimicrobial efficacy and tensile strength of the bioplastic were characterized. The bioplastic coating with high starch content (0.5% starch) enhanced WVTR, moisture absorption, water solubility and biodegradability due to the hydrophilic nature of starch and glycerol. Contact angle measurements showed the top surface with a value of 55.87° and the bottom surface with 45.66°. The side with the lower contact angle corresponded to increased water absorption and a higher WVTR. Peeled mangoes coated with starch-rich bioplastic exhibited accelerated water loss and improved retention of vitamin C. Incorporating CO-containing eugenol into the formulation provided antimicrobial effects, which decreased fruit fly oviposition during sun drying and as a result, minimized post-harvest losses. The biofilm also demonstrated adequate tensile strength and received favorable sensory evaluation scores. These properties make it a promising alternative to traditional plastics.

Sustainability spotlight

This research presents an innovative, biodegradable food coating and wrapper derived from cassava starch, agar and clove oil, which offer a dual-purpose solution reducing post-harvest losses and plastic pollution. By extending the shelf life of mangoes and providing antimicrobial protection against fruit fly infestation, the bioplastic directly supports SDG 12 (responsible consumption and production) and SDG 13 (climate action). Furthermore, its potential to replace single-use plastics contributes to SDG 14 (life below water) and SDG 15 (life on land) by mitigating microplastic contamination. The use of renewable, food-grade materials reinforces a circular economy, fostering SDG 9 (Industry, innovation and infrastructure) and promoting SDG 2 (zero hunger) through improved food preservation.

1 Introduction

Food security remains a critical global challenge, particularly in developing nations where access to sufficient, safe and nutritious food is threatened by high postharvest losses.[1,2] These losses not only reduce the availability of food but also represent a significant economic burden to producers. This further contributes to environmental stress through increased greenhouse gas emissions and waste generation. Globally, an estimated 20–50% of all fruits and vegetables produced are lost or wasted along the supply chain before ever reaching consumers.[3] These losses stem from a range of factors, including mechanical damage and biological threats such as pests and diseases, and the latter alone is responsible for over 40% of total fruit and vegetable losses.[4]

To reduce postharvest decay and extend the shelf life of fresh produce, some industries have begun applying synthetic plastic coatings directly onto fruits and vegetables.[5,6] These coatings act as protective barriers that limit moisture loss, slow oxidation and inhibit microbial growth. However, the widespread use of petroleum-based plastics has raised serious environmental concerns, particularly regarding their persistence in ecosystems and contribution to microplastic pollution.[7–9] In response, researchers in polymer science are actively developing biodegradable alternatives made from renewable natural resources such as starch, cellulose and plant-based proteins. These bioplastics can be formulated into edible coatings or cast into flexible films that serve the same protective functions as conventional plastics, while being compostable and non-toxic[10]. When applied to perishable, seasonal produce, these materials

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not only reduce spoilage but also offer a sustainable packaging solution that aligns with global efforts to mitigate plastic waste and promote environmentally responsible food preservation.[11]

Mangoes are regarded as a tropical seasonal fruit and are appreciated worldwide for their flavor, juicy flesh and nutritional values, such as vitamins A, C and B6, which help to enhance the body's immune system. The seasonality of mangoes makes them vulnerable to scarcity during their off-season period. The high perishability of mangoes leads to postharvest losses; therefore, the removal of water becomes a necessity to prolong the fruit's shelf life. Dehydration is one of the methods used for preserving mangoes, which can improve food security.[12] Traditionally, mangoes are dried outdoors in the sun, making them susceptible to fruit flies and other pathogens. The presence of fruit fly larvae in the mangoes also poses a biosecurity risk for many importing countries.[13] The use of synthetic chemicals to control pathogens has raised concerns related to human health and the environment. Consequently, finding solutions to prevent fruit fly infestations and extend the postharvest shelf life of mangoes has become a priority for the export market.

Edible coatings made from biodegradable polymers such as cellulose, starch, protein and chitosan offer a potential solution to the problems of food spoilage and single use plastics. These coatings form thin, consumable layers over food surfaces, acting as moisture and gas barriers while also serving as carriers for antimicrobial agents.[14] Starch is regarded as one of the most promising natural polymers to produce bioplastics due to its abundance, biodegradability and film-forming properties. As a naturally occurring polysaccharide found in plants like corn, cassava and potatoes, starch is a renewable and widely available resource. Its biodegradable nature allows starch-based bioplastics to break down naturally, reducing environmental pollution compared to conventional petroleum-based plastics. Furthermore, starch is non-toxic and safe for food-related applications, making it suitable for edible films and coatings. Although starch possesses many useful properties, its hydrophilic nature, brittleness and high water and oxygen permeability limit the formation of high quality bioplastics.[15] To address these shortcomings, starch is often blended with plasticizers like glycerol or other biopolymers such as chitosan and agar.[16,17] Agar is a polysaccharide mainly derived from seaweed, composed of two polysaccharides; agarose and agaropectin. The presence of agarose helps in gel formation, which makes it useful in developing effective bioplastics when combined with starch.[18]

An added advantage of this starch–agar formulation is its dual functionality: the same coating solution can be cast and dried into biodegradable plastic films. These films can be used as food-safe wrappers or packaging materials, offering a sustainable alternative to petroleum-based plastics. When dried, these films form flexible, transparent sheets with adequate tensile strength and barrier properties, making them ideal as eco-friendly packaging materials. Unlike conventional plastic wraps, these bioplastics are biodegradable, edible and compostable, making them suitable for short-term food storage, wrapping fast food items, or serving as inner liners for dried or minimally processed produce. This integrated approach not only addresses postharvest decay during drying and storage but also contributes to reducing plastic waste in the food supply chain.

Edible coatings based on biopolymers represent a sustainable alternative to conventional petroleum based plastic packaging.[19] Sourced from renewable materials, these coatings are biodegradable thereby reducing environmental impact and mitigating plastic waste. In addition, the production of biopolymer-based coatings typically requires lower energy inputs compared to conventional plastics and their non-toxic nature ensures environmentally safe disposal.[20,21] Although challenges remain in scaling up production and optimizing mechanical and sensory properties, edible coatings exemplify a sustainable approach that integrates food preservation with environmental responsibility, supporting circular economy initiatives and global efforts to reduce both plastic pollution and food waste.

In this study, edible food coating and wrappers were prepared using agar, clove oil (CO) and cassava starch. The incorporation of CO provided antimicrobial properties and acted as a natural insect repellent, addressing challenges such as fruit fly infestation and microbial spoilage of post-harvest produce during sun drying. Peeled mangoes immersed in the bioplastic solution and subjected to sun drying showed an improvement in post-harvest shelf life. The development of the bioplastic coating as a food wrapper presents a possible alternative to single-use plastics for food packaging. This research aims to provide an eco-friendly alternative to conventional plastics while enhancing food preservation and reducing plastic pollution.

2 Experimental

2.1 Materials

Day old harvested cassava tubers and mangoes were purchased from a local farmer (Suva, Fiji). Food grade agar and CO were purchased from a local supermarket (Suva, Fiji). All other chemicals were purchased from Sigma Aldrich and used as received.

2.2 Starch extraction

The cassava tubers were thoroughly washed to remove any soil, peeled and pulverized in a mechanical blender with distilled deionized water (DDW). The resulting mixture was transferred to a 1 L beaker and additional DDW was added and mixed before allowing the mixture to settle overnight. The solution was filtered through a double-layered cheesecloth and homogenized again with DDW before being left to settle for 24 h. This process was repeated twice to ensure maximum extraction. The starch obtained after decantation was freeze dried and stored in an airtight container for further analysis.

2.3 Chemical analysis of cassava starch

The total starch content of the cassava flour was measured using the Megazyme Total Starch Assay Protocol (amyloglucosidase/α-amylase method). Additionally, the ash content and the proportions of amylose and amylopectin in the cassava starch were quantified following AOAC methods, utilizing the Megazyme Amylose/Amylopectin Assay Kit (K-AMYL 06/18).

2.4 Preparation of the bio-edible coating solution

Cassava starch was mixed with 100 mL of DDW to prepare starch solutions with concentrations of 0.1, 0.3 and 0.5% (w/v). These solutions were gelatinized at 75 °C under continuous stirring at 600 rpm, followed by the addition of 2 mL of acetic acid. The temperature was increased to 90 °C and 2 g of agar was added, along with 1.2 mL of glycerol. The combined mixture was maintained at 90 °C for 30 min before being cooled to 40–50 °C while stirring continuously. At this stage, 1.5 mL of CO was incorporated into the mixture. This mixture was used for coating and bioplastic film preparation. For the coating, peeled mangoes were dipped into the mixture and allowed to dry on a wire rack. The mixture was maintained at around 50 °C to prevent the agar from solidifying. The coated mangoes were placed under direct sunlight for a period of nine days to facilitate drying and preservation. To create the bioplastic films, approximately 25 mL of the solution was poured into Petri dishes and air dried for 2 days and further dried in an oven at 40 °C for 24 h.

2.5 FTIR analysis

A PerkinElmer Spectrum Two Fourier Transform Infrared (FTIR) spectrophotometer equipped with an attenuated total reflectance accessory was used to obtain the IR spectra of the bioplastic films. An average of 64 scans at a resolution of 2 cm^{-1} were performed for each spectrum. Spectral analysis was done using the instrument's software. All spectra were normalized between 0 and 1 using eqn (1) for ease of comparison among spectra.[22]

$$\frac{(\text{Absorbance} - \text{absorbance}_{\min})}{(\text{Absorbance}_{\max} - \text{absorbance}_{\min})} \tag{1}$$

2.6 Tensile and thickness measurement

The thickness of the bioplastic films was measured using a micrometer (Mitutoyo Japan). Measurements were taken at five different locations on each film to determine the average thickness. The tensile properties of the bioplastic films were determined using a Shimadzu EZ Test Texture Analyzer equipped with a "General Tensile Jig Set". Samples were cut into rectangular strips measuring 5 cm × 1 cm for testing.

2.7 Contact angle determination

The surface contact angle was measured using the sessile drop method (ASTM D5946). The biofilm was cut into 3 × 6 cm pieces. Using a micropipette, 30 μL of water was dropped in the center of the biofilm. The side view image was photographed and analyzed using contact angle software. The contact angle is defined as the angle between a liquid and solid within a body of the liquid formed at the solid–liquid–gas interface. Measurements were performed on both sides of the biofilm. Five replicates were taken from different regions of each sample to ensure accuracy and reproducibility.

2.8 Water vapor transmission rate (WVTR)

The water vapor transmission rate (WVTR) of the bioplastic films was determined following a modified version of the method described in (ref. 23). The bioplastic films were cut into discs with an area of 4.91 cm^2 (A) and dried in an oven for 24 h at 105 °C. Boiling tubes were filled with 35 mL of DDW, ensuring a headspace of 5 cm remained. The bioplastic films were then carefully sealed onto the top of the boiling tubes using a suitable adhesive. The initial mass of the entire assembly (tube, water and film) was recorded and the assembly was then stored at room temperature for 24 h. The final mass was recorded after 24 h. The WVTR was calculated using eqn (2):

$$\text{WVTR}\ \left(\text{g m}^{-2}\ \text{h}^{-1}\right) = \frac{\left(w_\text{i} - w_\text{f}\right)}{24} \times A \times 10\,000 \tag{2}$$

where w_i and w_f refer to the initial and final weights and A is the area of the film.

2.9 Moisture content and water absorption

The moisture content of the bioplastic films was determined according to the AOAC 18th edition method.[24] Bioplastic films were cut into 1 × 1 cm, and their initial weight was recorded. The films were then dried in an oven at 105 °C for 24 h to obtain the final mass. The moisture content was calculated using eqn (3):

$$\text{Moisture content (\%)} = \frac{\text{initial weight} - \text{final weight}}{\text{initial weight}} \times 100 \tag{3}$$

The swelling of the films was measured following the ASTM D 570 method.[25] Dried films were immersed in 50 mL of DDW at room temperature for 24 h. The films were then removed, and excess surface water was immediately wiped off before weighing. The percentage of water absorbed was calculated using eqn (4):

$$\text{Swelling (\%)} = \frac{\text{final weight} - \text{initial weight}}{\text{initial weight}} \times 100 \tag{4}$$

2.10 Solubility in water

The water solubility of the bioplastics was measured using the method given in (ref. 26) with slight modifications. Bioplastic films were cut into 1 × 1 cm and dried in an oven at 105 °C for 24 h. The films were then soaked in 50 mL of DDW in centrifuge tubes and placed on a shaker for slow agitation (300 rpm) at room temperature for 24 h. The films were removed and dried again in an oven at 105 °C for 24 h to obtain the final mass. The solubility of the bioplastic films in water was calculated using eqn (5):

$$\text{Solubility in water } (\%) = \frac{\text{initial weight} - \text{final weight}}{\text{Initial weight}} \times 100 \tag{5}$$

2.11 Biodegradability test

The biodegradability test for the bioplastics was done similar to the method given in (ref. 27). The bioplastic films were cut into 1 × 1 cm pieces, dried in an oven for 24 h at 105 °C and accurately weighed. The films were buried in soil and weighed on a weekly basis. The biodegradation of the bioplastic films was calculated using eqn (6):

$$\text{Weight loss } (\%) = \frac{\text{initial weight} - \text{final weight}}{\text{initial weight}} \times 100 \tag{6}$$

2.12 Carbon dioxide evolution test

The aerobic biodegradation of the bioplastic films in soil was assessed by measuring carbon dioxide (CO_2) evolution, following the standard test method ASTM D 5988-03, with slight modifications.[28] The bioplastic films were dried in an oven for 24 h at 105 °C. 100 g of soil, with a moisture content of 54% and a pH of 7.60, was sieved to a particle size of 2 mm and placed in a biometer flask. 0.2 g of each bioplastic film sample was added to the flask. A control set-up was also prepared without any sample. A burette filled with charcoal was placed inside a rubber stopper, and the stopper was used to seal the biometer flask airtight. A tube connected to the biometer flask was inserted into an inverted 50 mL burette filled with DDW, which was submerged in a beaker partially filled with water. The burette readings were taken daily to monitor the displacement of water due to CO_2 evolution from the sample. The density of CO_2 at 27 °C was obtained from the literature and multiplied with the volume of the CO_2 released to determine the mass of CO_2. The percentage of CO_2 evolution per gram of the bioplastic film sample was calculated with respect to the number of days.

2.13 Weight loss of coated mangoes

The coated and uncoated mangoes were placed outside, under the sun. The weight of the mangoes was taken at three-day intervals for two weeks.

2.14 Determination of vitamin C

The vitamin C content of the dried mangoes was determined using High-Performance Liquid Chromatography (HPLC) according to the method described by Nyangena *et al.* (2019).[12] A 1 g sample of dried mangoes was extracted with 30 mL of 0.8% metaphosphoric acid. The mixture was then centrifuged at 5300 rpm for 20 min at 4 °C using a Thermo Scientific Heraeus Megafuge 16 Centrifuge. The supernatant was filtered through Whatman filter paper no. 4 into a 50 mL volumetric flask and the volume was adjusted to the mark with 0.8% metaphosphoric acid. After thorough mixing, approximately 1 mL of the solution was filtered through a 0.45 μm Millipore filter into an HPLC vial. A 20 μL aliquot of the extract was injected into the HPLC system.

Quantification was performed using a series of ascorbic acid standards (0.01 to 0.1 mg mL^{-1}) to generate a calibration curve. A UV detector set to 264 nm was used for detection. Separation was achieved using a C_{18} column (250 mm × 4.6 mm, 5 μm particle size) maintained at 25 °C in a column oven. The mobile phase consisted of 95% water, 5% methanol and 6 drops of 6 M sulfuric acid, with a flow rate of 1 mL min^{-1}. Vitamin C content was expressed as milligrams per 100 grams of dried mango.

2.15 Antimicrobial test

The bioplastic films containing 0.5% starch, glycerol and varying concentrations of CO were tested against *Escherichia coli* (*E. coli*) (bacteria) and *Rhizopus* (fungus). Broth prepared cultures of *E. coli* and *Rhizopus* were streaked on Mueller Hinton Agar and Potato Dextrose Agar plates, respectively. Gentamicin and ketoconazole were used as controls for bacterial and fungal strains, respectively. The bioplastic film was cut into circular shapes using a size 4 core borer and placed on bacterial and fungal streaked plates. The inoculated plates were incubated for 24 h and 48 h, respectively, at 37 °C and the zone of inhibition was measured. All aseptic techniques were followed and tests were conducted in triplicate.

2.16 Sensory evaluation

Sensory evaluation was conducted to assess the acceptability of the edible bioplastic. Sixty participants were randomly selected and provided with the bioplastic films. Each participant was given a questionnaire designed to evaluate five key sensory attributes: appearance, aroma, flavor, texture, and overall acceptability. Participants were instructed to rate each attribute on a 5-point hedonic scale, where 1 indicated "dislike extremely" and 5 indicated "like extremely". The responses were collected and analyzed to determine the general consumer perception of the edible bioplastic. The questionnaire is given as a SI (S1).

2.17 Statistical analysis

Statistical analysis was conducted using ANOVA: single factor ($p \leq 0.05$) in Microsoft Excel (Microsoft Corporation, Redmond, WA). Any significant differences were then examined. All experimental trials were conducted in triplicate and the average was taken. The error value was calculated based on the standard deviation of the triplicates for each concentration.

3 Results and discussion

3.1 Formulation of the edible bioplastic

From cassava flour, 73.4% of starch was extracted, which contained 19.07% amylose. The purity of the extracted starch was assessed based on the ash content, which was 0.15% ± 0.01. This low ash value indicated minimal inorganic residue, suggesting a high degree of purity consistent with food-grade starches.[29] The edible bioplastic was effectively formulated using food-grade cassava starch, agar, glycerol and CO, with their chemical structures illustrated in Fig. 1.

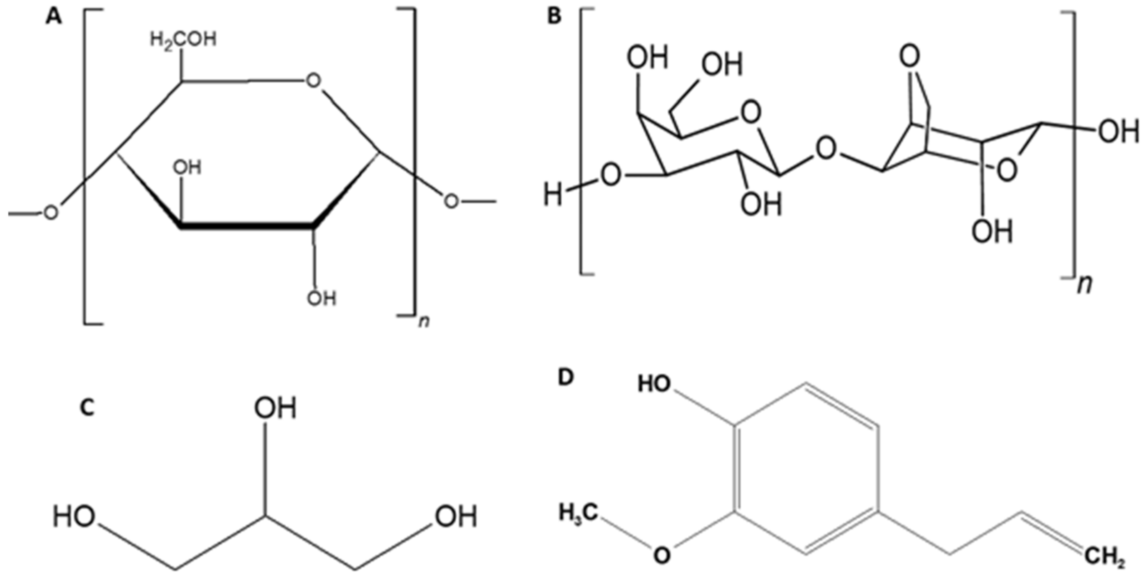

Fig. 1 The chemical structure of (A) starch, (B) agar, (C) glycerol and the most active compound, (D) eugenol, found in CO.

Cassava starch exhibited inadequate film forming ability; therefore, formulations were prepared with agar. Notably, the optimal amount of agar used for the bioplastic formulation was 2 g. The formulation only achieved suitable viscosity at an agar concentration of 2 g; otherwise, the resulting films were overly thin and structurally weak. The use of glycerol in the formulation of the bioplastic was anticipated to reduce brittleness and improve flexibility. The percentage composition of CO in the formulation was kept at 1.5% as this resulted in better antibacterial properties. Cloves are commonly used as a spice in cooking at low concentrations. Studies suggest that use of CO at low concentrations is generally considered safe for food applications.[30–32] The use of low dose CO in the formulation of the biofilm for mango preservation in this study is therefore considered to be acceptable, with minimal health risks to consumers. This formulation is environmentally sustainable, as it extends the shelf life of dried mangoes, generates no waste and results in a fully biodegradable, edible film.

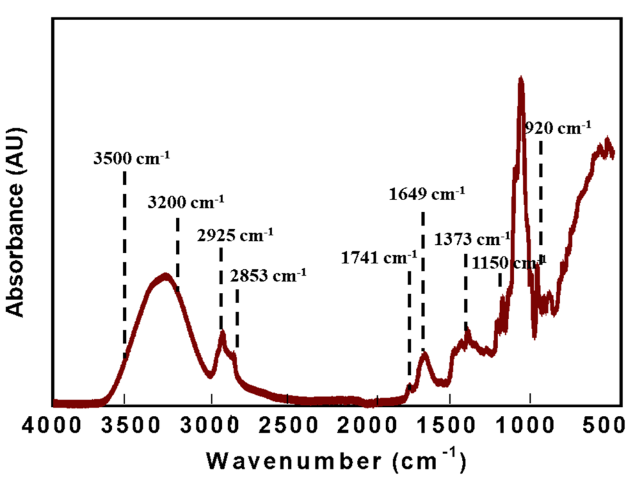

Fig. 2 Infrared spectrum of the 0.5% starch bioplastic.

3.2 FTIR analysis

Fig. 2 shows the FTIR spectrum of the biofilm containing 0.5% (w/v) of the starch/agar film. The broad band observed at 3500–3200 cm^{-1} is attributed to the vibration of the hydroxyl group present in starch, agar and glycerol. The presence of hydroxyl, carbonyl and ester groups, which are commonly found in biodegradable materials, supports the biodegradability of the bioplastic.[33,34] The bands between 2925 and 2853 cm^{-1} correspond to the CH stretch. The band at 1741 cm^{-1} is the stretching of the C–O group, with the C=C stretching vibration at 1649 cm^{-1}, together with the CH_2 bending band at 1373 cm^{-1}. The bands in the region between 1150 and 920 cm^{-1} are due to the C–O–C stretch and are attributed to the eugenol of CO. These peaks were not present in starch–agar–glycerol matrices and, therefore, provide strong evidence of successful incorporation of CO into the bioplastic. Bioplastics containing 0.1 and 0.3% starch showed similar FTIR spectra results as 0.5%; hence, their spectra have been omitted from Fig. 2 for clarity.

3.3 Water vapor transmission rate

The water vapor transmission rate (WVTR) is an important parameter used to assess the water vapor permeability of the bioplastics. As shown in Table 1, there was no significant difference ($p = 0.067$) in the WVTR with increasing starch content. For edible bioplastics, the WVTR has been reported to be between 1.34 and 3.42 g per m^2 per day[35] while that for starch and gelatin was 4.27 g per m^2 per day.[36] Starch chains are loosely packed and the gelatinization process disrupts the chain orientation, creating spaces among the molecules that facilitates water permeability[11] by creating small voids that retain

Table 1 The behavior of water with varying amounts of starch in the bioplastic

Bioplastic composition	WVTR (g per m^2 per day)	Moisture absorption (%)	Swelling (%)	Water solubility (%)
0% starch	1.77 ± 0.01	51 ± 2	53 ± 2	53 ± 2
0.1% starch	1.85 ± 0.01	61 ± 2	67 ± 2	67 ± 2
0.3% starch	1.93 ± 0.01	72 ± 2	75 ± 2	78 ± 2
0.5% starch	2.01 ± 0.01	84 ± 2	80 ± 2	87 ± 2

water molecules.[37] The WVTR obtained in our study aligns well with reported values for edible bioplastics and are much lower than reported starch/gelatin blends.[38] To reduce the WVTR for starch based bioplastics, hydrophobic materials such as oils or bentonite, which are capable of eliminating voids within the bioplastic structure, have been added.[23,39] Furthermore, incorporating essential oils into bioplastics also reduces the WVTR due to their lipidic or hydrophobic properties.[39,40] Our work has shown that by incorporating agar, glycerol and CO in specific ratios, the WVTR can be controlled. Incorporating 1.5% CO in the formulation did not considerably affect the WVTR probably due to the low concentration that may not have imparted sufficient hydrophobicity. A similar finding was reported in (ref. 41) where CO itself did not influence WVTR, but the high starch content was responsible for the elevated WVTR.

An increased WVTR with a higher starch content can be beneficial, especially during sun drying of fruits, as it promotes more efficient moisture removal, thereby accelerating the drying process. This rapid drying helps prevent microbial growth and enzymatic spoilage, ultimately preserving the fruit's quality. Additionally, shorter drying times reduce exposure to environmental factors that can degrade sensitive nutrients like vitamin C, ensuring better retention of nutritional value. Overall, starch-enriched edible coatings that enhance the WVTR provide an effective strategy for improving fruit preservation by enabling faster, safer drying while maintaining both quality and nutritional integrity.

3.4 Contact angle, moisture absorption, swelling and water solubility

Wettability is an important surface property of biofilms, as it determines the extent to which a liquid can spread or be absorbed on the film surface.[42] It is commonly evaluated by water contact angle measurements, where lower contact angles indicate hydrophilicity and higher angles indicate hydrophobicity as shown in Fig. 3.[43]

Table 2 presents the contact angle measurements of the bioplastics after placing a water droplet on the surface. The contact angle of the biofilms increased with increasing starch content and was significantly different ($p < 0.001$) for each blend system. This indicated that the water droplet spread less, which suggested reduced interactions between the water and the bioplastic surface. Although higher starch levels are typically associated with increased hydrophilicity due to the presence of more hydroxyl groups leading to a decrease in contact angle, the observed increase may be attributed to the formation of self-hydrogen bonds among hydroxyl groups in the dry state.[44] Over time, the contact angle gradually decreased.

Furthermore, contact angle measurements were conducted on both sides of the biofilms with CO. As the coating solution dried within the Petri dish, one side was exposed to air while the bottom remained in contact with the glass surface. The analysis showed a significant difference ($p < 0.001$) between the two surfaces of the biofilm with clove oil. Notable differences in wettability between the two sides were observed. For the biofilm containing 0.5% starch, the surface exposed to air exhibited an average contact angle of 55.87°, indicating increased hydrophobicity, whereas the opposite side showed a lower contact angle of 45.66°, reflecting a more hydrophilic nature. This asymmetric wettability arises from differences in polarity between the two sides: the air-facing surface becomes enriched with hydrophobic, non-polar (CO) due to its low density, while the opposite side remains more hydrophilic, facilitating vapor diffusion.[45] When applied to a wet mango and exposed to sunlight during drying, the hydrophilic side, sticking to the mango side, readily absorbs water from the fruit, driven by the water concentration gradient between the mango and the coating plus the micro voids formed within the film matrix by the hydroxyl groups present in starch, enhancing water uptake and diffusion pathways for vapor transport.[46] Conversely, the air-facing side of the film containing CO reduces the contact area and surface tension of the escaping water molecules, thereby accelerating evaporation.[47,48] This dual functionality where one side resists moisture penetration but promotes water evaporation, and the other is more permeable and prone to absorption significantly influences the overall barrier properties of the coating.

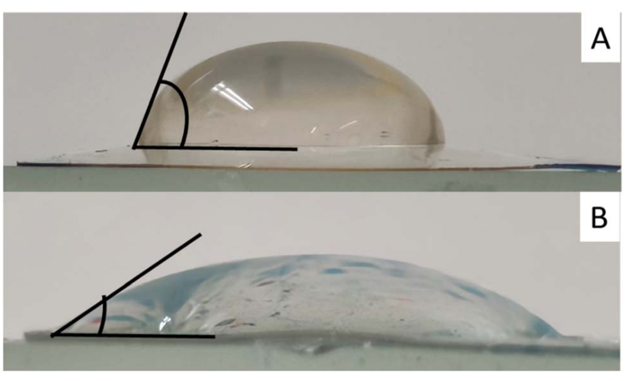

Fig. 3 The contact angle of water on (A) the air facing surface and (B) the bottom side of the biofilm.

Table 2 Contact angle (θ) of the bioplastic film with and without clove oil

	Bioplastic with clove oil		Bioplastic without clove oil
	Upper	Lower	
0% starch	27.60 ± 4.21	24.55 ± 1.40	26.83 ± 1.32
0.1% starch	39.72 ± 4.77	28.23 ± 2.37	30.45 ± 2.34
0.3% starch	51.00 ± 2.54	36.92 ± 2.36	36.50 ± 3.75
0.5% starch	55.87 ± 1.73	45.66 ± 3.85	41.43 ± 2.73

The moisture absorption, swelling and water solubility of the bioplastic increased as the starch content increased as shown in Table 1. The bioplastic with 0.5% starch exhibited the highest moisture absorption (84 ± 2%), swelling (80 ± 2%) and solubility (87 ± 2%), whereas the control sample (0% starch) had sufficiently lower values. It was found that the starch content in the bioplastic had a significant effect on the moisture absorption ($p < 0.001$), swelling ($p = 0.0001$) and water solubility ($p < 0.001$). The presence of hydroxyl groups in starch, agar and glycerol facilitated the formation of hydrogen bonds, enabling the matrix to retain water. This confirms a direct correlation between starch content and water interactions, consistent with findings in previous studies.[37,49]

Furthermore, the literature indicates that glycerol, a commonly used plasticizer, enhances water solubility due to its hydrophilic nature and multiple hydroxyl groups.[11,50] Plasticizers weaken intermolecular interactions between polymer chains, increasing free volume within the matrix. This structural modification facilitates water penetration, thereby elevating the solubility and absorption capacity of the bioplastic.[11]

3.5 Biodegradation

The formulated bioplastic also exhibited biodegradability. Fig. 4 shows the % weight loss of the bioplastics with varying amounts of starch over three weeks measured at 1 week intervals when buried in soil.

The % weight loss of the bioplastics increased with time but varied for different compositions. Bioplastics with a higher starch content showed greater weight loss. A significant difference ($p = 0.033$) was observed in the mean % weight loss across different starch contents in the bioplastics. Several factors such as polarity, surface area, molecular weight, water absorption and microorganisms affect the biodegradation rate.[11] The hydroxyl groups in starch, agar and glycerol enhanced water absorption and hydrolysis, which, combined with microbial activity, facilitated the biodegradation of the bioplastic.[18] Microorganisms metabolize the bioplastics, breaking it down into carbon dioxide and water. Glycerol and starch contribute to the degradation process by enhancing water absorption, creating a favorable environment for microbial activity. As water serves as a medium for most microorganisms, its penetration into the biofilm accelerates biodegradation, particularly when placed in soil.

Additionally, glycerol weakens the hydrogen bonds (which increases the water resistance and flexibility of the bioplastic) therefore increasing the biodegradation of the bioplastic.[50] Fig. 5 shows the biodegraded bioplastics containing 0.5% starch up to week 3. The formation of perforations on the surface of the biofilms allowed the diffusion of enzymes and oxygen, increasing the biodegradation process as shown in Fig. 3A–D. The presence of carbonyl (C=O), hydroxyl (O–H) and ester (COOH) groups in the bioplastic confirmed by FTIR suggests that biodegradation of the bioplastic would occur at a rapid pace.[34] Therefore, an increase in starch content in the bioplastic increased water absorption consequently, accelerating the biodegradation of the bioplastic films.

The anticipated life cycle of each bioplastic differs depending on the type of bioplastic and on the end-of-life scenarios

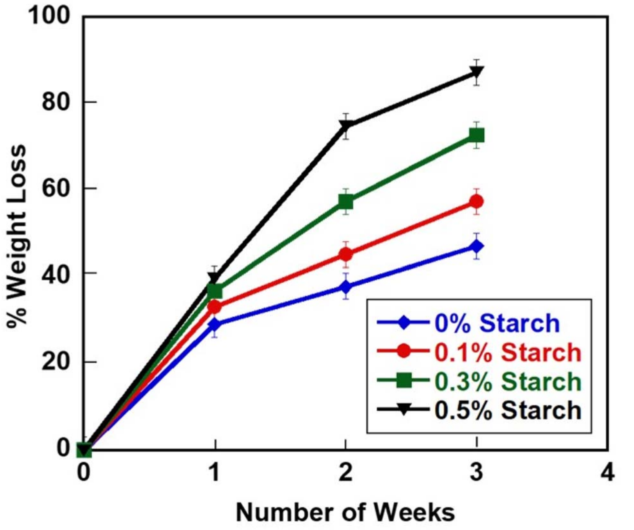

Fig. 4 The % weight loss of bioplastic films with time when buried in soil.

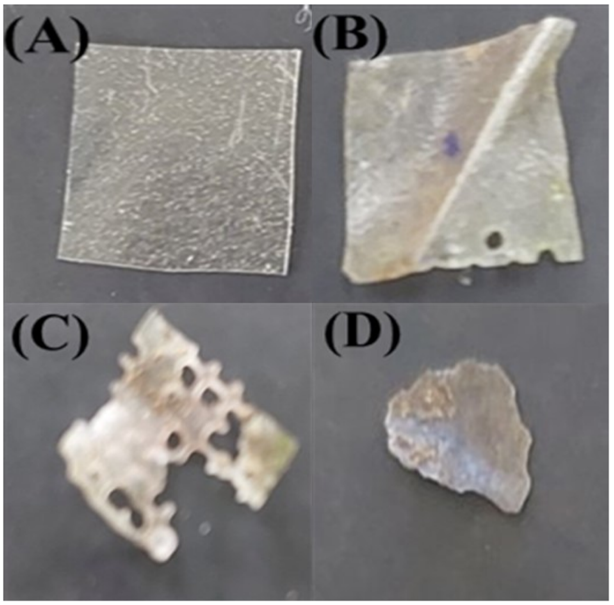

Fig. 5 The biodegradation of 0.5% bioplastic at: (A) (0 week), (B) (1 week), (C) (2 weeks) and (D) (3 weeks).

according to the available waste management systems of each country. The biodegradation observed for the starch–agar–CO films was compared with results reported in the literature on other biodegradable films, providing a meaningful reference point. With regard to biodegradation in the soil, polylactic acid (PLA) degraded ~10% over 98 days, PLA/sisal fiber composites degraded less than 60% in the same period, starch/chitosan films degraded ~80% in 14 days, polybutylene succinate/starch blends degraded ~24.4% in 28 days, polycaprolactone/starch blends degraded ~37% over 270 days and polyhydroxyalkanoates degraded 40–50% in 15 days.[51,52] In comparison, the 0.5% starch–agar–clove oil films prepared in this study degraded ~80% within just 21 days, demonstrating rapid soil biodegradability and establishing them as a highly promising environmentally friendly bioplastic alternative.

3.6 Carbon dioxide evolution

Fig. 6 presents the evolution of CO_2 per gram of the sample during biodegradation. The evolution of CO_2 increased with increasing starch content in the biofilms. A significant difference ($p < 0.01$) was observed in the cumulative CO_2 released after 7 days in biofilms containing different starch contents. This result is aligned well with the biodegradation test, supporting the biodegradability of the edible bioplastic coating. Starch films are prone to microbial growth, which can lead to elevated CO_2 production.[49]

3.7 Tensile strength and thickness

The thickness of the bioplastic ranged from 0.12 to 0.18 mm and could be controlled by adjusting the volume of the mixture poured into the Petri dish. The tensile strength of the bioplastic films was evaluated under dry and wet conditions. Fig. 7 shows a typical tensile stress–strain curve for the formulated bioplastic. The mechanical properties of the bioplastic were compared with those of edible rice paper. Under dry conditions, the rice paper exhibited the highest tensile force (50.35 N),

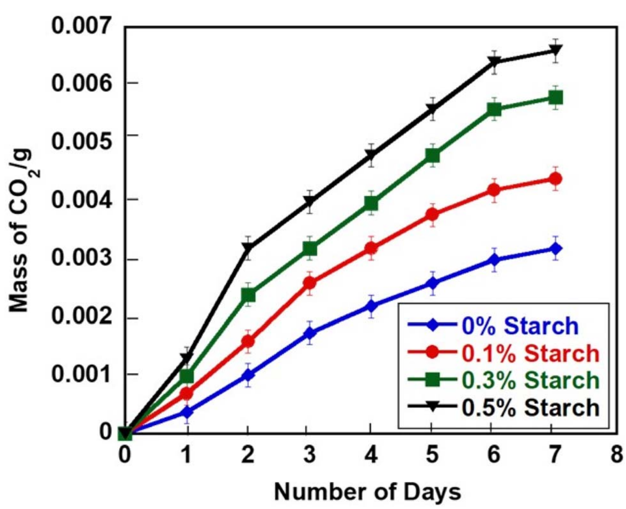

Fig. 6 CO_2 evolution from bioplastics with time.

followed by the bioplastic with 0.5% starch (14.96 N), 0.3% starch (12.7 N), 0.1% starch (15.94 N) and 0% starch (12.09 N). Increasing the starch content did not show any considerable difference ($p = 0.068$) in the tensile strength. The starch helps to form strong films by forming a more cohesive interaction through hydrogen bonding.

The molecular interactions between starch and agar affected the film's mechanical properties, with low agar levels leading to a marked decrease in tensile strength. Conversely, higher agar concentrations resulted in thicker films that negatively impacted sensory acceptance. To optimize cost-effectiveness without compromising consumer appeal, the concentration of CO was maintained at a relatively low level, as small variations in CO did not significantly alter the tensile strength of the coatings. These findings are consistent with those reported in (ref. 4).

Since the starch content did not have a substantial effect on the tensile strength, only the 0.5% starch biofilm was tested under wet conditions. Under wet conditions, the bioplastics showed a significant reduction in tensile strength due to water absorption weakening the intermolecular forces as shown in Fig. 7C. The swollen bioplastic samples had an average tensile force of 3.04 N, a decline from their dry-state performance. Similarly, rice paper experienced a drastic reduction from 50.35 N (dry) (Fig. 7A) to 0.72 N (wet) (Fig. 7B), indicating its susceptibility to moisture and weakening of the intermolecular forces. The reduction in tensile strength aligns with the hydrophilic nature of starch and glycerol, which enhances water absorption and weakens structural integrity. Practically, for edible films, it is expected that the tensile strength of the biofilm should decrease in the wet state to allow ease of chewing if applied as a food coating.

The results from this study also indicate that starch-based bioplastics exhibit promising tensile strength, particularly under dry conditions, making them suitable for applications such as short-term food packaging and wrapping. The wet films experienced a reduction in strength but remained more flexible for wrapping wet food and were easier to handle. This makes the bioplastic a more practical alternative for applications requiring some moisture resistance. However, for chewing purpose, the bioplastics were slightly stronger compared to the rice paper. The bioplastic could offer a viable, sustainable alternative to conventional plastic packaging while maintaining adequate mechanical performance.

The long-term mechanical stability of the bioplastics, assessed through 12 month storage at ambient temperature, was maintained. Re-evaluation of tensile strength revealed no substantial degradation, confirming their structural integrity. This, coupled with the observed reduction in tensile strength under wet conditions (indicating desirable flexibility and chewability for edible coatings), demonstrates excellent performance across a range of practical conditions.

3.8 Analysis of antimicrobial properties

Essential oils are volatile liquids extracted from aromatic plants, known for their insect-repelling properties, that help

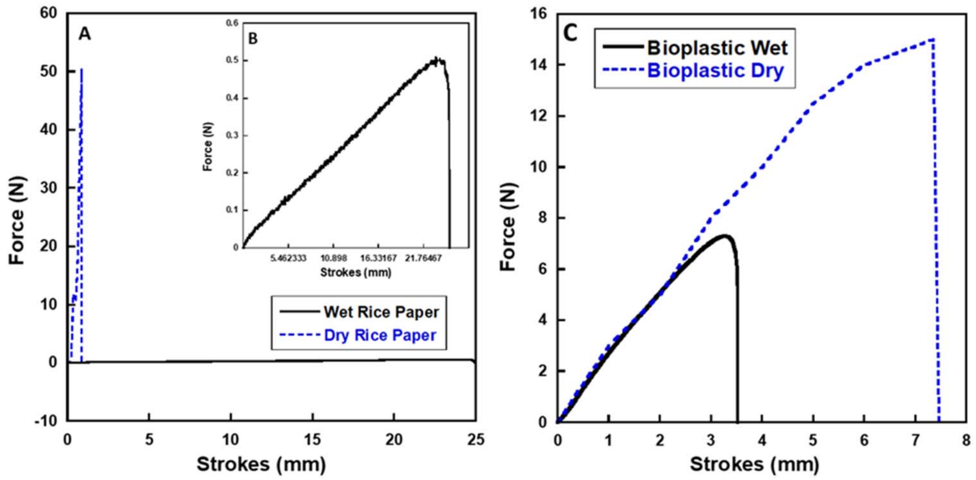

Fig. 7 Tensile results for (A) dry rice paper, (B) wet rice paper, and (C) wet and dry bioplastic.

control spoilage both before and after harvest.[53] CO extracted from the *Syzygium aromaticum* plant has a high amount of eugenol which has anti-inflammatory and analgesic properties.[54] Additionally, the eugenol in CO irritates fruit flies and distracts them, while also possessing antimicrobial properties effective against several microorganisms.[53]

Fig. 8 shows the zone of inhibition against (A) *Escherichia coli* (Gram-negative bacteria) and (B) *Rhizopus* (fungus), respectively, for the bioplastic containing agar, glycerol, 0.5% starch and varying concentrations of CO ranging from 0.2 to 2.0%. The antimicrobial activity of the bioplastic films was assessed using the zone of inhibition as the primary quantitative endpoint, providing a direct measure of the extent of microbial growth suppression around the film samples. There was no zone of inhibition in starch and agar bioplastic without CO. The zone of inhibition increased with increasing amount of CO in the bioplastic. The minimum inhibitory concentration (MIC) in this study was determined as the lowest concentration of CO incorporated into the films that consistently produced a clear and measurable inhibition zone against both the tested bacterial and fungal strains. The MIC of CO against *E. coli* was 1.5% (Fig. 8A) and 1% for *Rhizopus* (Fig. 8B). Therefore, 1.5% CO was selected to be the optimum concentration to be incorporated into the bioplastic films, aiming to provide broad-spectrum antimicrobial protection.

Eugenol, which is a phenolic compound found in CO, is able to disintegrate the complex cell wall structure of *E. coli* which comprises outer membrane proteins, lipopolysaccharides and a peptidoglycan layer.[55] Hence, CO can become a promising antimicrobial agent in inhibiting the growth of some foodborne pathogens in the bioplastics.

3.9 Bioplastic sensory evaluation

The sensory evaluation of the bioplastic is presented in Table 3.

According to Table 3, the high overall score of 4.90 out of 5 for the 0.5% starch formulation reflects a strong consumer

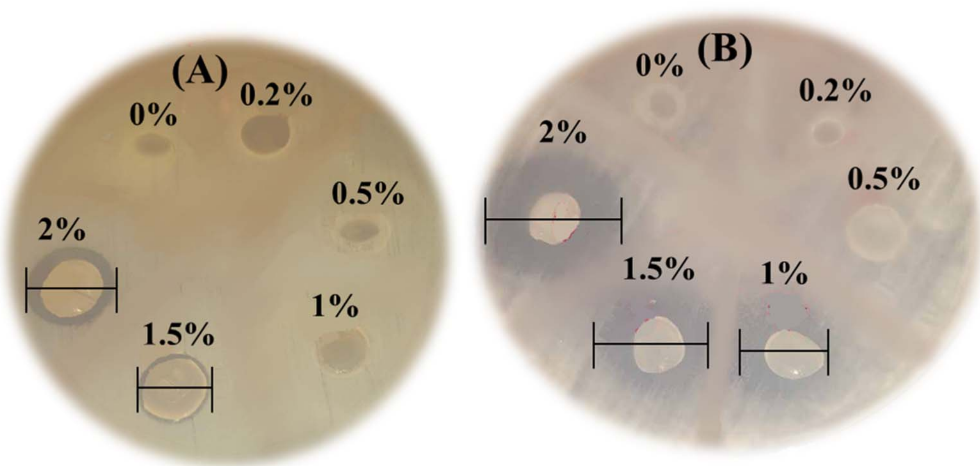

Fig. 8 The minimum inhibitory concentration (A) antibacterial and (B) antifungal tests conducted on *E. coli* and *Rhizopus*, respectively, using varying % of CO.

Table 3 Sensory evaluation results of the edible bioplastic coating

Bioplastic composition	Appearance	Flavor	Texture	Aroma	Overall
0% starch	4.04	3.10	3.87	3.95	3.74
0.1% starch	4.51	3.54	4.18	4.73	4.24
0.3% starch	4.57	4.58	4.38	4.76	4.57
0.5% starch	4.82	4.92	4.93	4.94	4.90

preference, especially regarding flavor and texture. These results align with previous studies indicating that a higher starch content enhances palatability and results in a more uniform film structure, while the incorporation of essential oils positively influences aroma and perceived freshness.[34,56] The consistently high aroma scores across all samples suggest that, beyond providing antimicrobial benefits, CO also enhances sensory appeal. Additionally, the improved appearance ratings with increased starch concentration are likely due to greater transparency and structural uniformity of the films, which meet consumer expectations for visually attractive edible materials. The favorable flavor ratings further indicate that the combined use of starch and CO offers a mild yet appealing taste, with clove oil possibly imparting a subtle spicy note.

From a practical standpoint, sensory acceptability is crucial for the successful commercialization of edible coatings and films. Consumers tend to be cautious about edible packaging unless it delivers a neutral or pleasant flavor and texture; thus, the near-maximum scores achieved by the 0.5% starch formulation suggest that this material can be seamlessly integrated into food products without negatively affecting consumer perception. The bioplastic's ability to simultaneously deliver functional performance such as moisture control, antimicrobial properties and high sensory acceptance underscores its valuable potential for industry adoption.

An added advantage of the bioplastic coating is that the solution can be poured and dried as sheets which can be effectively used as wrappers. Fig. 9 shows a wrapped sandwich with the bioplastic film. The transparency of the bioplastic allowed for visual inspection. These characteristics are essential considerations within the framework of Hazard Analysis and Critical Control Points (HACCP) to ensure food safety and quality. The shelf life of the bioplastic placed in sealed polyethylene bags was monitored at 1 month intervals for 12 months and was found to be unaffected.

Fig. 9 A sandwich wrapped using the bioplastic containing 0.5% starch.

This bioplastic wrapper is suited for fast foods such as sandwiches, wraps, burgers and fries which are not stored for long periods to avoid moisture absorption. The ecofriendly and biodegradable qualities of the bioplastic make it suitable both as a food coating and as a wrapper, serving a dual purpose.

3.10 Effectiveness of the bioplastic coating

Fig. 10A shows the mango coated with CO, (B) mango coated without CO and (C) uncoated mangoes. The mangoes coated with CO showed no signs of infestation or spoilage after 2 days of drying. The strong aroma of CO, primarily due to its high eugenol content, acted as a natural repellent against fruit flies, larvae, mosquitoes, ants and other pests.[53] In tropical countries, fruit flies are among the most common pests responsible for fruit spoilage during post-harvest. The genus *Drosophila*, belonging to the family *Drosophilidae*, is widely recognized as a major group of fruit flies.[57] When sliced mangoes were left uncovered, their moisture and fragrance created an ideal environment for the oviposition of fruit flies. Approximately 48 hours later, adult fruit flies emerge, with females becoming sexually mature and capable of laying eggs soon after.[58] This leads to about 40% of post-harvest spoilage during sun drying.[59] The incorporation of CO in the coating created an unfavorable environment for fruit fly oviposition and reduced microbial contamination. Uncoated and coatings without CO exhibited the presence of maggots and fungal growth after 2 days of sun drying. It has been also reported that a 1.5% CEO-chitosan coating on strawberries effectively inhibited mold and yeast growth, prolonging preservation.[54]

3.11 Analysis of water loss in mango samples

Coated and uncoated mango samples were sun-dried to improve preservation and post-harvest decay.[60] The water loss from the mango samples was monitored by weight as shown in Table 4. Significant difference ($p < 0.05$) was observed in the mean % weight loss between the uncoated sample and the starch-coated sample.

As the starch content in the coating increased, the mango samples experienced greater weight loss due to water loss.[54] The coating absorbs water from the mango and loses it to the atmosphere due to the difference in the hydrophilicity of the two sides of the coating and this has been explained in Section 3.4.

3.12 Analysis of vitamin C

Mangoes are highly valued for their rich nutrient content, particularly Vitamin C, a heat-sensitive micronutrient.[12] In this study, monitoring vitamin C levels became a critical aspect of assessing the preservation efficacy of the bioplastic coating. Table 4 shows the comparative analysis of vitamin C content between fresh and sun-dried mangos.

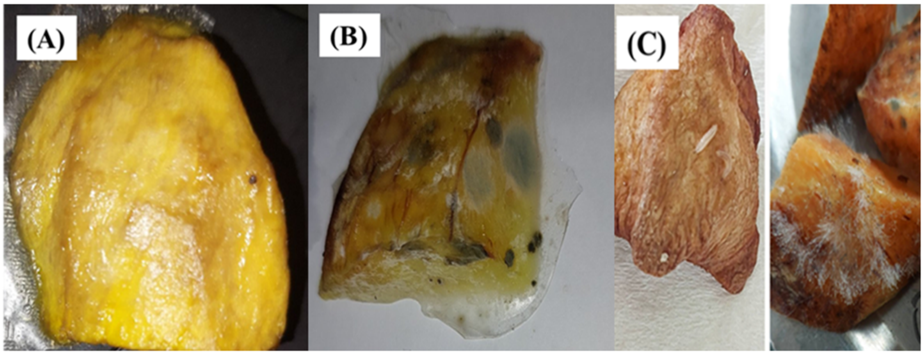

Fig. 10 Mango samples: (A) coated with CO, (B) coated without CO and (C) uncoated.

Table 4 Percentage weight loss of mango samples coated with various types of coatings as a function of time and vitamin C content of coated and sundried mango samples

	Weight loss (%)				
Samples	3 days	6 days	9 days	Samples	Vitamin C (mg/100 g)
				Fresh mango	40 ± 2
Uncoated	66 ± 2	74 ± 2	79 ± 2	Uncoated sun dried	8 ± 2
0% starch	69 ± 2	76 ± 2	87 ± 2	0% starch sun dried	11 ± 2
0.1% starch	73 ± 2	79 ± 2	89 ± 2	0.1% starch sun dried	11 ± 2
0.3% starch	79 ± 2	82 ± 2	92 ± 2	0.3% starch sun dried	11 ± 2
0.5% starch	82 ± 2	90 ± 2	95 ± 2	0.5% starch sun dried	11 ± 2

In fresh mangoes, the vitamin C content was found to be around 40 mg/100 g. This value is within the range reported in the literature between 9.79 and 186 mg/100 g.[12,61] The variation in the vitamin C content can be correlated to the variety of mango, post-harvest handling, geographical distribution, stage of growth and degree of ripeness.[62]

The vitamin C content decreased considerably in sun dried mangoes compared to fresh mangoes and is likely due to the heat sensitivity of vitamin C. Uncoated sun-dried mangoes contain considerable moisture content, which enhances enzymatic reactions leading to loss of vitamins through oxidation in the presence of oxygen.[12] Despite there being no significant difference ($p > 0.05$) in vitamin C levels between coated and uncoated mangoes, the weight loss analysis indicated that the bioplastic coating, due to its increased WVTR from the hydrophilic nature of the coating, accelerated moisture removal from the mangoes. This enhanced the drying rate, achieved without compromising the vitamin C content, suggesting that the bioplastic coating could offer a viable strategy for mango preservation, particularly through sun-drying and improving postharvest decay. Beyond mangoes, other tropical and subtropical fruits including papaya, banana, pineapple and guava could similarly benefit from coatings with a higher WVTR during drying, since these fruits are also highly perishable and often preserved through dehydration to extend shelf life and reduce postharvest losses.

Compared to commercially used coatings, which are often synthetic, non-edible and dependent on chemical preservatives, starch/agar-based edible coatings incorporated with CO provide several functional and sustainability advantages.[63–65] Conventional postharvest treatments with synthetic waxes and chemical fungicides have long been applied to reduce decay and extend fruit shelf life. However, these treatments rely on preservatives such as benzoic acid, sodium benzoate, sorbic acid, potassium sorbate and propionic acid, whose rapid diffusion into the food matrix lowers their surface concentration and reduces antimicrobial effectiveness over time. Continuous use of such chemicals has also raised concerns regarding human health, environmental contamination, and the emergence of resistant pathogenic strains.[66] In contrast, starch/agar coatings enriched with CO form a biodegradable, fully edible matrix capable of providing a semi-permeable barrier against moisture and oxygen while simultaneously delivering the controlled release of natural antimicrobial and antioxidant compounds. This dual functionality both suppresses microbial growth and delays oxidative degradation, thereby extending shelf life without the drawbacks of chemical residues. Conventional coating materials often exhibit poor mechanical strength and high sensitivity to moisture, whereas the starch/agar-based coating developed in this study demonstrated enhanced mechanical strength and more favorable water resistance. Collectively, these advantages highlight starch/agar/CO coatings as a superior alternative to conventional synthetic coatings for postharvest preservation.

4 Conclusion

Eco-friendly, edible food coatings and wrappers made from bioplastics were formulated using agar, CO and cassava starch which exhibited several advantageous properties. Higher starch content in the bioplastic enhanced its water solubility, water absorption, moisture uptake, WVTR, biodegradability, carbon dioxide evolution and sensory quality. The addition of agar and glycerol improved tensile strength in the dry state.

Mangoes coated with the bioplastic exhibited increased water loss during drying, indicating its potential to facilitate faster dehydration. The sensory evaluation confirmed that the 0.5% starch bioplastic had the highest overall acceptability, making it the most preferred formulation. Moreover, the inclusion of CO provided antimicrobial activity, effectively inhibiting bacterial and fungal growth, while also repelling fruit flies and preventing oviposition and maggot infestation. These attributes highlight the bioplastic's suitability for food coating and wrapping applications. The combined mechanical and environmental benefits, along with favorable sensory acceptance, demonstrate the bioplastic's potential for widespread adoption.

Future research could focus on enhancing the starch–agar–CO coatings by integrating additional active agents, such as natural antioxidants, probiotics, or complementary essential oils, to further improve their antimicrobial and preservative properties. Moreover, extending the application of these coatings beyond mangoes to a variety of fruits and vegetables, including papaya, pineapple, banana, guava and leafy greens, would offer valuable insights into the formulation's versatility across different produce types. Additionally, investigating the performance of these coatings under various storage conditions, such as refrigeration or controlled atmosphere environments, would be beneficial for optimizing their effectiveness.

The findings from this research offer a promising approach in improving post-harvest decay of mangoes, while also mitigating plastic pollution and its environmental impacts, including waste management challenges, climate change and microplastic contamination in the food chain. Further development and scaling of this technology could significantly reduce plastic waste and post-harvest food losses, supporting a more sustainable and environmentally responsible future.

Author contributions

David Rohindra: project administration, supervision and funding acquisition, conceptualization, data analysis, writing – review and editing. Reeha Sharma: investigation, data analysis, writing the first draft. Roselyn Lata: data analysis, writing – review and editing. Tejesvi Patel: data analysis, writing – review and editing.

Conflicts of interest

The authors declare no conflict of interest.

Data availability

All data, models, and/or code that support the findings of this study are available with in the article.

Supplementary information: questionnaire used for sensory evaluation. See DOI: https://doi.org/10.1039/d5fb00409h.

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