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ORIGINAL PAPER

Sustainable biodegradable packaging films based on cationic potato-peel starch/broccoli with edible inks

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Abstract

This study presents the development of a novel eco-friendly biodegradable films based on cationized potato-peel starch incorporated with broccoli-derived bioactive compounds extracted using deep eutectic solvents (DES). Fourier transform infrared spectroscopy (FT-IR), Differential scanning calorimetry (DSC), Scanning electron microscopy (SEM), Water vapor (WVTR) and oxygen permeabilities (OTR) were used to fully characterize the prepared edible films. The effect of adding broccoli extract to starch films was evaluated as bioactive compounds through antioxidant and antimicrobial examination. Addition of broccoli extract to modified starch films improves resistance, moisture and gas transmission and mechanical properties. FTIR and DSC techniques confirmed extra amorphous and heterogeneous structure of the composite as a result of successive addition of broccoli extract into the cationic potato-peel starch. SEM images revealed that increasing the percentage of the incorporated broccoli into cationic potato-peel starch resulted in a more amorphous surface structure which facilitated the printability of the film and its quality. The developed films improved mechanical flexibility (E%), reduced WVTR and OTR with 28%, 20%, 34% respectively, alongside enhanced antioxidant and antimicrobial properties. The overall migration tests were conducted according to EN 1186-5 and EN 1186-4 with range from 0.1 to 1.3 dm^3. These tests confirmed the fully compliance of the tested material with European food contact safety requirements. These findings will highly empower the development of new packaging materials with extended shelf life and eco-friendly nature as edible packaging flexible films.

Keywords Sustainable packaging · Biodegradable packaging film · Potato-peel starch · Edible printing inks · Agro-waste valorization

Introduction

Many advantages of plastic applications like remarkable safety and hygiene properties for food packaging, good water and chemical resistance, outstanding electrical and thermal insulation properties are stated. On the other hand, some disadvantages of plastic applications have been reported as environmentally harmful through pollution for living in their surroundings. Global great increment of plastic consumption reveals alarming environmental impact. Nearly 400 million metric tons of plastic packaging materials are produced annually, which greatly responsible to the incredible degradation in our environment [1–3]. Moreover, biopolymers, a prospective novel solution in the packaging industry, represent promising solutions to overcome the disadvantageous of plastic industry. These polymers serve as environmental alternatives that protect wildlife by decreasing plastic pollution, while also complying with the global environmental regulations [4, 5]. In addition, providing environmentally friendly global challenges, these biopolymer-based packaging introduce many examples as a safe, low-cost, non-toxic, and edible materials. Starch presents as one of the most exceptional popular substitutes to plastic as a biopolymer. It reduces the expected environmental and health hazards in packaging applications [6]. Moreover, it is

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one of the most important natural resources of great availability, as it can be easily obtained from various agricultural and industrial by-products [7, 8].

Furthermore, starch is the second most available natural biopolymers after cellulose [9]. It provides many environmental solutions to plastic drawbacks. Besides, it offers various advantages, including fundamental biodegradability, great processability and excellent biocompatibility with other materials and acceptable barrier resistance to both carbon dioxide and oxygen. Starch-based materials represent outstanding example for use as edible coatings in several food packaging manufactures [10]. In addition to high encouraging applications as edible coatings, starch meet the dual goals of using renewable resources and providing end-of-life advantages (biodegradation or ingestion). Recent studies have characterized starch-based edible packaging in terms of rheology, thermal behavior, film formation, biodegradability, barrier properties, and mechanical performance [7–9].

Many inexpensive renewable resources in our universe, potato peel is one of the most economical sources of starch, a naturally biodegradable byproduct of potato processing and is very rich of starch [11]. The necessity of using agricultural byproduct to produce biodegradable packaging films is to decrease carbon footprint and fulfill the severe environmental regulations. Consequently, there is increasing demand for starch-based films as the most sustainable packaging option in the food industry [12, 13]. Meanwhile, there are some drawbacks of using starch in packaging like its increased hydrophilicity, decreased water vapor barrier properties and decreased mechanical strength. Therefore, starch modification is greatly convenient to sort these drawbacks out.

Modified starch has attracted extensive attention greater than natural starch due to its outstanding chemical and physical properties. Several modification methods of starch including chemical, physical and enzymatic treatments to change their properties [14, 15]. Among these, chemical methods are the most convenient approaches that used in starch modification. Cationic starch, an important example of starch derivative, has chains of positively charged groups. It is of increased water solubility and outstanding film-forming properties that successfully used in biopolymer-based films preparation [16]. This in turn, increases its value in foods preservation.

Starch-based materials, among the most famous edible coatings, have emerged as effective solution for vegetables and fresh fruits preservation. Generally, edible inks are preferred over conventional inks due to the toxicity of conventional inks and many numerous problems they cause in the packaging industry, such as migration into food which causes contamination of packaging films, difficult and costly recycling. It was reported that edible coatings, natural alternative to traditional preservation methods are of great versatility. Examples of edible coatings include waxes, chitosan, beeswax, and carboxymethyl-cellulose. This versatility allows coatings to preserve nutrients such as antioxidants, flavoring agents and antimicrobials to great extent [17]. Moreover, these coatings have high ability to decrease water loss, increase product gloss, control respiration rates and finally have outstanding ability to suppress microbial growth throughout postharvest storage [12, 13]. Besides, edible inks, food-grade colorant designed for printing onto edible surfaces, are of great importance in packaging industry. They reduce production cost and manufacturing cycle and hence decreases the expected environmental and health hazards in packaging applications and edible printing [18]. Moreover, these edible inks fulfil the human safety requirements of consumption as they are of Food and Drug Administration (FDA)- approval food dyes [19].

From all the points mentioned above, it is clear that food industry has become one of the largest sources of waste in modern time, causing health problems worldwide. The increasing amounts of waste generated and accumulated by the vegetable industry have disrupted biodiversity and significantly impacted human health and climate change. Therefore, wastes results from agricultural food industries are of great importance to requires the global attention, mainly because of the pollution and economic mismanagement it causes. Hence, the reduction of generated wastes is critical from economic value point of view [20, 21]. Besides, this waste reduction can be enhanced by incorporation of agri-food loss in industry as for example packaging films. Moreover, some waste materials contain high levels of bioactives that can be extracted to be the good starting material for developing a novel biodegradable packaging film of antibacterial and antioxidant effect.

Broccoli by-products, a source of bioactive extracts from waste, is edible plant and exhibits various antimicrobial and antioxidant effects with health benefits and antioxidant effects of broccoli [22]. In this research, we propose to develop eco-friendly biodegradable packaging films based on a tailored cationic potato-peel starch was presented as a valorized agro-waste stream combined with broccoli (vegetable by-product) substrates, onto which edible inks are printed.

Experimental

Materials and methods

Potato peels were collected from local food processing facilities and washed carefully before starch extraction. Broccoli residues, including stems and leaves, were obtained from agricultural waste. All chemical reagents used for starch cationization, including sodium trimetaphosphate (STMP) and sodium tripolyphosphate (STPP), are of analytical grade, Sigma-Aldrich, Germany. 2,2- diphenyl-l-picrylhydrazyl (DPPH) is Sigma-Aldrich product, Germany. *Staphylococcus aureus*, *Pseudomonas aeruginosa*, *Candida albicans* and *Aspergillus niger* were kindly obtained from Microbiology Dep., Al-Azhar University, Egypt.

Extraction and cationization of potato-peel starch

Potato-peel starch was extracted using a standard wet milling process, followed by drying and sieving. The extracted starch was then subjected to cationization using STMP and STPP in an alkaline medium to introduce positive charges onto the starch chains. The degree of substitution (DS) of cationic starch was determined using titration methods to confirm the efficiency of the modification.

Preparation of biodegradable films

Dispersing cationic potato-peel starch in distilled water was performed to prepare biodegradable films, followed by the incorporation of broccoli-derived fibers. The film genic solution was stirred continuously at 70 °C to ensure homogeneous dispersion. Casting the solution onto petri dishes for drying at controlled temperature and humidity conditions. This was followed by peeling off the films and storing in desiccators before characterization.

Preparation of soluble starch from potatoes waste

Starch was extracted from potato-peel waste following the procedure outlined by Torres et al. [23]. The potato peels were cut into small pieces and mixed with deionized water in a 1:2 solid-to-liquid ratio under continuous stirring (70 min, 220 rpm). The resulting mixture was then passed through an 80 µm sieve, and the retained residue was rinsed with deionized water to recover any residual starch. The filtrate was subsequently refrigerated for one day to facilitate starch sedimentation. The separated starch was collected, dried in a blast drying oven at 40 °C for 48 h, and stored in a desiccator till further use. Starch yield from potato-peel waste averaged 14.4%, with 19.29% amylose content.

The deep eutectic solvent (DES) was prepared according to the methodology described by Almeida et al. [24]. Briefly, the DES method was established according to the following steps, The components are stirred, with in a 1:2 molar ratio, and temperature from50–60 °C until a homogeneous, transparent liquid is formed. After that, the addition of water was approx. (e.g., 50 wt% or more) to optimize the viscosity and extraction efficiency for phenolic compounds from plant matrices. This method is chosen for being an eco-friendly and effective alternative to conventional solvents for extracting bioactive.

Preparation extract of broccoli

Washing broccoli stems and leaves with distilled water, cutting into small pieces of 0.5 cm × 0.5 cm, freeze drying, grinding, and sieving with 80 mesh sieve to obtain broccoli powder with small particle size of less than 177 µm; adding aqueous extraction solvent, performing ultrasonic-assisted extraction at 50 °C for 11 min. with 40% amplitude with Branson sonicator, England, 70 kHz.

Preparation soluble starch/broccoli extract films

Three ratios of extract were added with 1, 3, and 5% to soluble starch solutions with 10% w/w glycerol as plasticizer. The mixture was applied under vigorous stirring by ultra-homogenizer at 13,000 rpm for 6 min followed by ultrasonication at 30% amplitude for 7 min. The solutions were casting in Teflon petri dishes and drying overnight in air refreshing oven at 60 °C.

Characterization techniques

Mechanical properties

The mechanical properties of the prepared films, including tensile strength (TS) and elongation at break (E%), were evaluated by a universal testing machine Z005 TN/TH, Zwick, Germany. The measurements were performed in accordance with ASTM D882 standards to assess film flexibility and durability.

Water vapor and oxygen permeabilities

Water vapor transmission rate (WVTR) was performed by GBI W303 (B) Water Vapor Permeability Analyzer, China, by means of the cup method at temperature (38 °C) and (90% RH) in accordance with ASTM E96, ISO 2528, ASTM D1653, TAPPI T464, DIN 53122-1 and JIS Z0208) standards. Also, the oxygen transmission rate (OTR) under condition (23 °C and 50% RH) was determined by N530 Gas Permeability Analyzer (China). The test was carried out in accordance with ISO2556-2001, GB/T1038-2000 and ASTM D1434-82(2003) standards.

Fourier transform infrared spectroscopy (FTIR)

FTIR analysis was performed to prove the chemical modification of the starch and the interactions between starch and broccoli-derived components by Alpha II Bruker (Germany) spectrometer, a horizontal ATR device. Existence of functional groups indicative of cationization and bioactive compounds was identified through spectral analysis.

Scanning electron microscopy (SEM)

SEM (Quanta FEG 250), Japan, was applied to study the surface morphology of the films, providing insights into their microstructure, homogeneity, and fiber dispersion within the polymer matrix.

Differential scanning calorimetry (DSC)

DSC analysis was conducted to examine the thermal behavior of the films by DSC 8500 from PerkinElmer, USA, including glass transition temperature (T_g) and thermal stability, which are critical for evaluating their heat resistance and processing conditions.

Overall migration test

Several worldwide rules are official with regard to migration of certain chemicals such as heavy metals, additives and degradation products. These compounds may have unpleasant taste, smell, or supposed adverse results to customers. All details of overall migration (OM) simulants and settings are described in EU Regulation Nr. 10/2011. It is expressed by the total milligrams of substance lost from 1 decimeter square surface (mg/dm^2). Generally, all results of overall migration were calculated in accordance to (EN 1186-5) [25].

Antimicrobial activity of the tested extracts

Capability of the tested materials to prevent the microbial proliferation was examined against the standard bacterial and fungal Pathogen strains, Gram-positive bacterium (*Staphylococcus aureus*) Gram-negative bacterium (*Pseudomonas aeruginosa*), unicellular fungi (*Candida albicans*) and multicellular fungi (*Aspergillus niger*). Pre-activation of pathogens were performed by inoculating in the Nutrient broth medium for one day, at 37 °C for bacterial strains, whereas fungal strains were inoculating in Potato Dextrose Broth (PDB) medium for 48 h. at 28 °C with shaking condition to activate and obtaining a heavy spore size. Screening of the examined compounds was carried out at fixed disc dimension (3 × 3 mm) using Mueller Hinton Broth (MHB) via turbidometry technique for every microbial pathogen excepting A. niger, this was discussed by Ragab et al. [26]. Furthermore, the evaluation of tested compounds to inhibition the microbial growth was also evaluated at the significant concentration by Agar-well diffusion method using Mueller Hinton Agar (MHA). Obtained results were evaluated depending on the inhibition zone diameter (mm) as suggested by Clinical and Laboratory Standards Institute (CLSI) guidelines. Furthermore, examined materials were assessed in compared to the standard antibacterial and antifungal agents as a positive control, whereas DMSO was treated as a negative control [27].

Consequently, the Minimum Inhibition Concentration (MIC) test for the sample's efficiency was prepared in contradiction of each microbial pathogen. Regarding this aspect, the broth microdilution method was applied depending on CLSI, meanwhile an identified weight of the potent sample was dissolved in DMSO to prepare a stock solution. Every targeted material was diluted using the broth medium (nutrient for bacterial strains or potato dextrose for fungal strains) to get the chosen concentration (25–250 µg/ml). The calculation of the MIC value for every targeted compound was determined as the lowest concentration of every sample which generated a minimum number of colony-forming units (CFU) related to the reference samples [26].

Antioxidant activity of the tested extracts

Determination of antioxidant activity of the tested extracts was performed using DPPH free radical scavenging assay as described in previous research work [26]. DPPH Radical scavenging activity freshly prepared (0.004% w/v) methanol solution of 2,2- diphenyl-l-picrylhydrazyl (DPPH) radical was prepared and stored at 10 °C in the dark. 500 µl of each extracted solution was transferred into the tested tube and mixed with 500 µl of DPPH solution [28]. Instantly, measurements of absorbance were determined using a UV-visible spectrophotometer (Agilent Cary-100, Germany). Absorbance decrement at 515 nm was continuously recorded, with data reported at 1 min. Intervals until the absorbance stabilized (16 min). DPPH radical absorbance without antioxidant (control). Also, the measurements of reference compound ascorbic acid were performed. DPPH radical percentage inhibition (PI) was calculated according to the formula: PI = [{(AC-AT)/AC} x 100]

AC= Absorbance of the control at t = 0 min and AT= absorbance of the sample + DPPH at t = 16 min. The IC50 value of every antioxidant assay was determined and expressed as µl/mL.

Results and discussion

Mechanical properties

Mechanical properties of starch-based composite films incorporated increased concentrations of broccoli extract (1–5%) reveals a clear trend in tensile behavior and flexibility. The blank sample, pure starch film, (S1) exhibited a tensile strength (TS) of 14.2 ± 0.4 MPa and an elongation at break (E) of 28.4 ± 1.1%, representing a relatively rigid and brittle biopolymer matrix.

The mechanical properties of the starch-based composite films were expressed as mean ± standard deviation, revealing a clear influence of broccoli extract incorporation on the tensile behavior of the films. Table 1 shows the gradual addition of broccoli extract, a progressive decrement in tensile strength and increment in elongation were observed. At 1% extract (S2), TS slightly decreased to 13.7 ± 0.5 MPa, while elongation increased to 32.1 ± 1.3%, indicating the initial plasticizing or compatibilizing effect of bioactive components from the extract. Increasing the broccoli extract to 3% (S3) and 5% (S4) further reduced TS to 11.9 ± 0.6 and 10.4 ± 1.4 MPa, respectively, while elongation rose to 34.2 ± 1.4 and 35.4 ± 1.6%. One-way ANOVA analysis revealed statistically significant differences among the films with different broccoli extract concentrations. Tensile strength, the ANOVA test showed a highly significant effect ($F = 35.65$, $p < 0.001$). Similarly, elongation at break exhibited significant differences between samples ($F = 15.24$, $p \approx 0.0015$), confirming that the incorporation of broccoli extract significantly influences the mechanical performance of the starch-based films. The relatively small standard deviation values indicate high reproducibility of the mechanical measurements and suggest a uniform and homogeneous structure of the prepared films.

This inverse relationship between strength and flexibility suggests that the broccoli extract acts as a natural plasticizer, weakening intermolecular hydrogen bonding among starch chains and increasing molecular mobility. The reduction in tensile strength may result from partial disruption of the polymer network or dilution of the starch matrix by low-molecular-weight phenolic and carbohydrate compounds.

Overall, the results demonstrate that moderate addition of broccoli extract enhances film flexibility and ductility at the expense of tensile strength. Such behavior can be advantageous for applications requiring improved film elasticity, foldability, or conformability, particularly in biodegradable packaging systems. Tensile strength and elongation of potato starch-chitosan/Eugenol were enhanced by addition of 6% Eugenol [12] this is in good agreement with the present results.

Table 1 Mechanical properties (tensile strength and elongation at break) of starch and starch with different ratios of broccoli extract

Nr.	Sample	Composites	Thickness, µm	TS, MPa	E, %
1	**(S1)**	Starch	70 ± 3	14.2 ± 0.4	28.4 ± 1.1
2	**(S2)**	Starch/1% broccoli	67 ± 2	13.7 ± 0.5	32.1 ± 1.3
3	**(S3)**	Starch/3% broccoli	72 ± 5	11.9 ± 0.6	34.2 ± 1.4
4	**(S4)**	Starch/5% broccoli	71 ± 3	10.4 ± 0.5	35.4 ± 1.6

Water vapor and oxygen permeabilities

Barrier performance of starch-based films was estimated through measurements of WVTR and OTR. The pure starch film (S1) exhibited relatively high permeability values, with a WVTR of 4.317 ± 0.21 g·mm/m²·day·kPa and an OTR of 2107 ± 11.1 cm³/m²·day·kPa, signifying the inherent hydrophilic and porous nature of pure starch.

The addition of broccoli extract resulted in a progressive decrease in both water vapor transmission rate (WVTR) and oxygen transmission rate (OTR) values as the extract concentration increased. For 1% broccoli extract (S2), WVTR and OTR decreased to 4.011 ± 0.32 and 1877 ± 14.2, respectively. Further addition to 3% (S3) and 5% (S4) resulted in a significant reduction to 3.674 ± 0.28 and 3.455 ± 0.35 g·mm/m²·day·kPa for WVTR, 1503 ± 17.5 and 1374 ± 13.2 cm³/m²·day·kPa for OTR, respectively, as shown in Table 2.

This consistent decline in permeability demonstrates that the broccoli extract effectively enhances the film's barrier properties. This improvement could be related to several concurrent mechanisms: (1) hydrogen bonds formation among phenolic compounds from the extract and hydroxyl groups of starch, leading to a denser polymer network; (2) partial filling of microvoids by extract molecules, reducing free volume and diffusion pathways; and (3) increased hydrophobicity due to nonpolar constituents in the extract, which hinders water and oxygen diffusion. WVTR showed significant variation between films ($p \approx 0.06$), while OTR exhibited a highly significant difference ($p < 0.001$) with increasing broccoli extract content. Similarly, overall migration results showed significant differences among food simulants ($p < 0.001$). The relatively small standard deviation values with indicate good experimental reproducibility and homogeneity of the prepared films.

The results confirm that broccoli extract addition to starch films improves resistance to moisture and gas transmission, with the most pronounced effect observed at 5% extract loading. These results indicating that broccoli extract is an effective natural modifier to improve the barrier performance of starch-based biopolymer films, making them more suitable for active food packaging applications which necessitate continued shelf life and enhanced protection against oxidation and moisture. WVTR of cassava starch, glycerol, sodium bentonite and cinnamon oil was notably enhanced by increasing cinnamon oil content up to 5% [9] this coincides with the present results.

Table 2 Water vapor and oxygen permeabilities of starch and starch with different ratios of broccoli extract

Nr.	Sample	WVTR, g.mm/m^2.day.kPa	OTR, cm^3/m^2.day.kPa
1	**(S1)**	4.317 ± 0.21	2107 ± 11.1
2	**(S2)**	4.011 ± 0.32	1877 ± 14.2
3	**(S3)**	3.674 ± 0.28	1503 ± 17.5
4	**(S4)**	3.455 ± 0.35	1374 ± 13.2

Fourier transform infrared spectroscopy (FTIR)

FTIR spectrum of the soluble starch matrix derived from potato waste and incorporated with different percentages of broccoli extract is shown in Fig. 1. It demonstrates the characteristic carbohydrate and phenolic vibrational features. The broad band observed around 3200–3600 cm^{-1} corresponds to O–H stretching vibrations, which increase in width and intensity as the broccoli extract content rises, indicating enhanced hydrogen bonding interactions between starch hydroxyls and phenolic hydroxyl groups from the extract. C–H stretching bands near 2850–2950 cm^{-1} show minor intensity variations, attributed to additional aliphatic groups introduced by extract constituents.

New or intensified absorptions between 1700 and 1740 cm^{-1} and 1600–1650 cm^{-1} indicate C=O and aromatic C=C or amide I vibrations, confirming the incorporation of phenolics and proteinaceous materials from broccoli into the starch matrix. The fingerprint region (1200–900 cm^{-1}) displays the typical C–O, C–O–C, and C–OH stretching bands of starch; however, changes in their relative intensities and minor peak shifts suggest partial disruption of starch crystalline order and formation of new hydrogen-bonding networks. Such modifications imply effective molecular interaction and compatibilization between the two components.

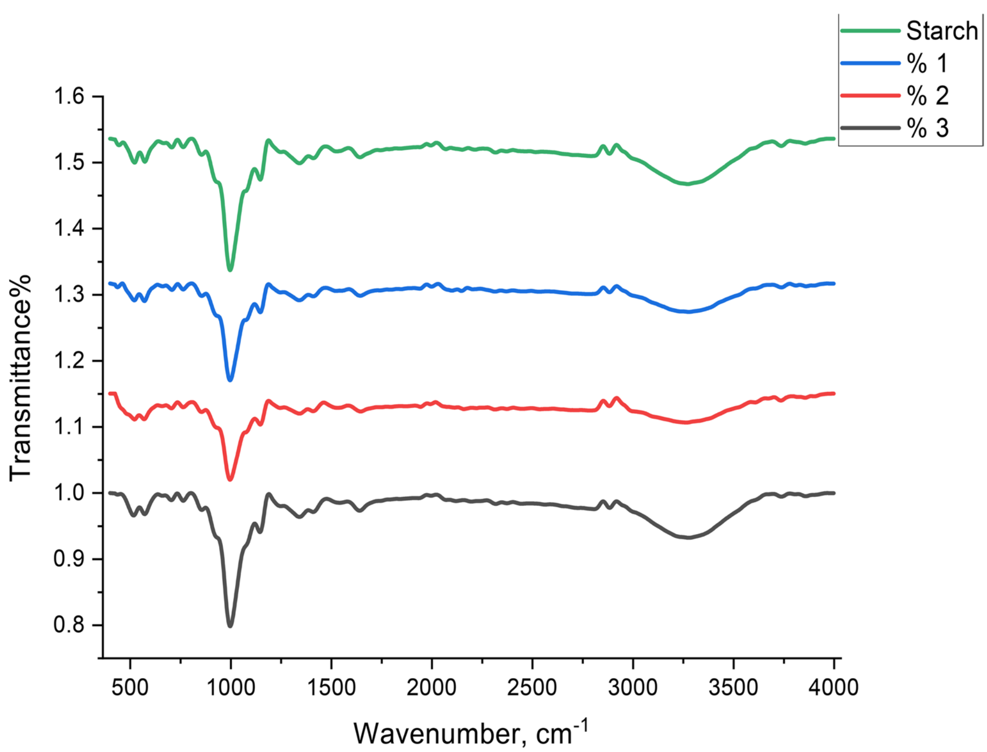
Overall, the FTIR results confirm successful integration of broccoli extract into the starch matrix through hydrogen bonding and secondary interactions, leading to increased amorphous and heterogeneous structure. These spectral changes indicating an enhanced intermolecular compatibility, which may beneficially influence the physical, thermal, and barrier properties of resulting biopolymer films.

Scanning electron microscopy (SEM)

SEM is an important technique to analyze materials' surface morphology at the microscopic level. In this discussion, the SEM images of starch films were integrated with different concentrations of broccoli extract (1%, 3%, and 5%) in samples S2, S3, and S4, respectively. Functional and structural properties of starch films were significantly influenced by the incorporation of bioactive compounds from broccoli extract.

Sample S1 serves as the control starch film, providing a baseline for comparison against samples S2, S3, and S4, which incorporate varying concentrations of broccoli extract. AS shown in Fig. 2, SEM images of S1 reveal a smooth and homogeneous surface morphology, indicative of a well-formed starch matrix without any added bioactive compounds. This uniformity suggests optimal film formation and structural integrity, making it an effective control for evaluating the impact of broccoli extract on film properties. The absence of extract allows for a clear understanding of how the introduction of natural additives influences surface characteristics and potential applications in biodegradable packaging. Further analysis of S1's properties will highlight the enhancements brought about by the integration of broccoli extract in subsequent samples.

Fig. 1 FTIR spectrum of starch with variable ratios of broccoli extract



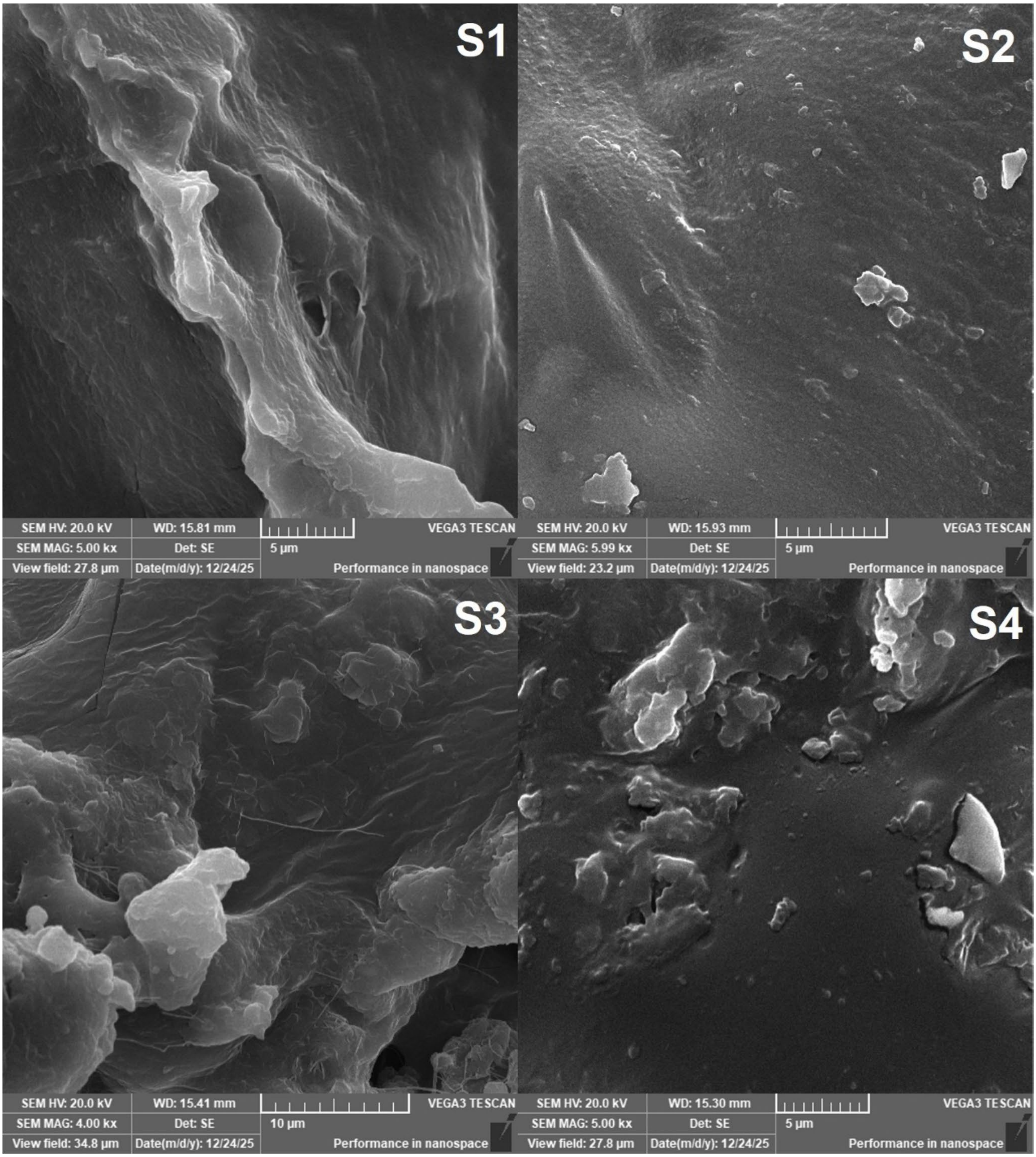


Fig. 2 Topographic images of starch film with and without different ratios of broccoli extract

The SEM images of S2 reveal a relatively smooth surface with minimal irregularities. The low concentration of broccoli extract likely enhances the film's integrity without significantly altering the starch matrix. Additionally, the broccoli extract appears uniformly dispersed, indicating good compatibility with the starch polymer. SEM images show an increase in surface roughness compared to S2. The higher concentration of broccoli extract may lead to agglomeration, resulting in uneven surface texture. The presence of broccoli extract at this concentration enhanced the formation of pores and cavities, which could influence the film's permeability and moisture absorption properties. Previous research by Kola et al. demonstrated that such modifications could improve the functional properties of biodegradable films [29].

S4 depict a significantly rougher surface with pronounced features like pores and cracks. This suggests a higher degree of extract integration, potentially leading to film brittleness. The structural changes observed in S4 may compromise the mechanical strength of the film. Different studies by Apostolidis et al. declared that excessive incorporation of fillers can lead to reduced tensile strength and elasticity [30].

The incorporation of broccoli extract not only alters the surface morphology but also enhances the functional properties for example antioxidant activity, moisture barrier properties, and biodegradability of starch films. Deshmukh et al. have indicated that natural extracts can enhance the antioxidant characters of films, and hence increase the films suitability for food packaging applications [31].

The varying concentrations of broccoli extract may be designed to verify specific functional requirements in food packaging, that increased barrier properties and antioxidant activities are crucial. starch films with varying concentrations of broccoli extract highlights significant morphological changes that correlate with the varying ratios of the extract. While lower concentrations lead to smoother surfaces conducive to mechanical integrity, higher concentrations introduce surface roughness and porosity that could affect the film's physical properties. Further research focusing on the mechanical and barrier properties, alongside a comprehensive analysis of the antioxidant activity of these films, is warranted to fully understand their potential applications.

Differential scanning calorimetry (DSC)

DSC thermogram of the pure starch with different ratios of Broccoli extract is Cleary shown in Fig. 3. The pure starch sample (S1) shows a characteristic broad endothermic region extending from approximately 40–150 °C, which is typical for native starch systems undergoing moisture evaporation, relaxation of amorphous domains, and partial disruption of crystalline structures. The relatively large negative heat-flow signal observed for S1 indicates a higher degree of crystalline–amorphous transitions and energy consumption associated with the breakdown of native starch granule organization. This behaviour reflects the strong internal hydrogen bonding within the starch matrix, which requires significant thermal input to disrupt.

When broccoli extract was incorporated at 1% (S2), the DSC curve exhibited a noticeable shift and the appearance of a more pronounced endothermic dip around 140–150 °C. This deeper endothermic feature suggests that small amounts of extract components (such as phenolics, organic acids, and small bioactive molecules) interact selectively with starch chains, forming localized ordered complexes that require higher thermal energy for disruption. At this concentration, the extract appears to partially reinforce or reorganize specific starch domains rather than fully plasticize the matrix, leading to a distinct thermal event not present in the pure starch sample.

Increased uniform and less intense thermal profile were observed by increasing the extract concentration to 3% (S3). The sharp endothermic transition observed in S2 becomes less defined, and the overall heat-flow curve shifts upward. This behaviour suggests that higher concentrations of broccoli extract begin to act predominantly as plasticizing and disordering agents, interfering with intermolecular

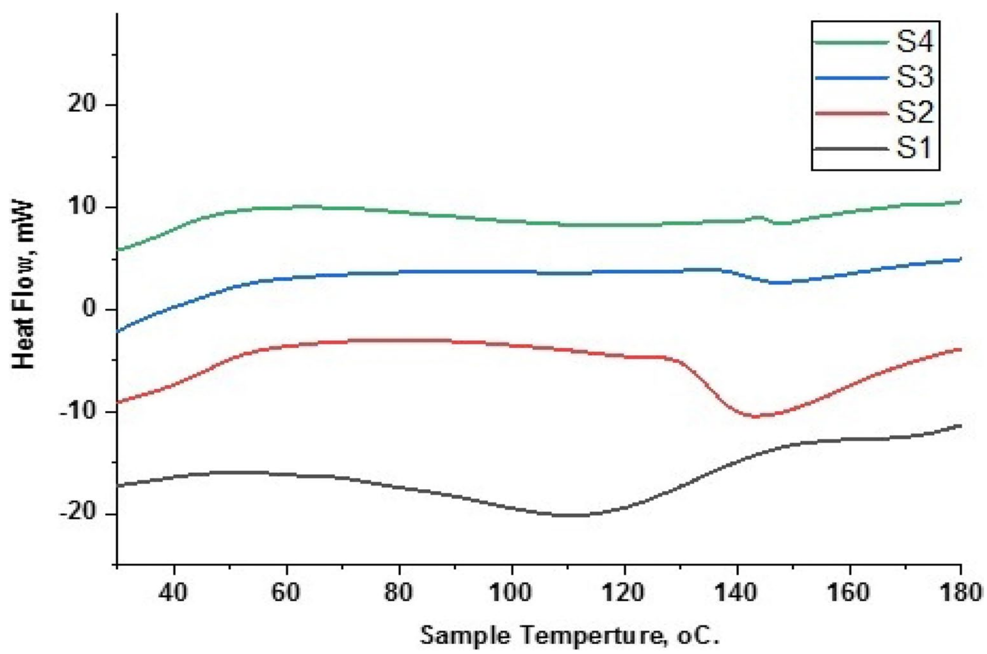


Fig. 3 DSC thermograms of starch with variable ratios of broccoli extract

hydrogen bonding among starch chains. Consequently, the crystalline regions become less stable and require less energy to disrupt, producing a smoother and lower-enthalpy transition across the heating range.

At the highest extract content (5%, S4), the DSC thermogram presents the most elevated baseline and the smoothest transition profile, with minimal sharp endothermic features. This indicates a substantial reduction in starch crystallinity and a higher proportion of amorphous, extract-modified domains. The extract likely enhances molecular mobility through hydrogen-bond interactions with starch hydroxyl groups, leading to a more flexible matrix and reduced cooperative thermal transitions. The progressive smoothing of thermal events and reduction in enthalpy from S2 to S4 clearly demonstrate that increasing broccoli extract concentration disrupts the native starch network and enhances its amorphous character.

Overall, the DSC results reveal a concentration-dependent effect of broccoli extract on the structural organization of starch. Low extract levels (S2) induce specific interactions that create distinct thermal domains, whereas higher concentrations (S3 and S4) lead to increasing plasticization, reduced crystallinity, and broader, less pronounced thermal transitions. These findings confirm that broccoli extract significantly alters the thermal behaviour of starch matrices, with implications for processing stability, film flexibility, and functional performance in biodegradable packaging applications.

Overall migration test

Overall migration tests were conducted according to EN 1186-5 and EN 1186-4 standards using food simulants A (ethanol 10%), B (acetic acid 3%), C (ethanol 20%), and D2 (vegetable oil or isooctane substitute) to represent aqueous, acidic, alcoholic, and fatty food contact conditions, respectively as presented in Table 3.

Across all simulants, the measured overall migration values varying from 0.1 to 1.4 mg/dm², with mean values of 0.4, 0.8, 1.3, and 0.1 mg/dm² for simulants respectively A, B, C, and D2. All obtained results are significantly below the regulatory limit of 10 mg/dm² detailed by EU Commission Regulation (EU) No 10/2011 and related standards, indicating excellent overall migration resistance of the tested material.

Table 3 The overall migration from starch/5% extract broccoli

Method Replicates	EN-1186-5 (simulant A) mg/dm^2	EN-1186-5 (simulant B) mg/dm^2	EN-1186-5 (simulant C) mg/dm^2	EN-1186-4 (simulant D_2) mg/dm^2
1	0.3	0.9	1.2	0.1
2	0.5	0.7	1.3	0.1
3	0.4	0.8	1.4	0.1
Mean result	**0.4 ± 0.1**	**0.8 ± 0.1**	**1.3 ± 0.1**	**0.1 ± 0.0**
Limit	**10.0**	**10.0**	**10.0**	**10.0**

The slightly higher migration observed in simulant C suggests a modest tendency of extractable components to migrate in moderately alcoholic environments, likely due to increased polymer–solvent interactions or partial solubilization of low-molecular-weight compounds. However, the very low migration values across all simulants demonstrate the material's chemical stability and inertness under diverse food contact conditions.

The overall migration values obtained for different food simulants were statistically analyzed using one-way ANOVA. The results demonstrated significant differences among the simulants ($p < 0.05$). Migration values followed the order C > B > A > D2, with mean values of 1.30 ± 0.1, 0.80 ± 0.1, 0.40 ± 0.1, and 0.10 ± 0.00 mg/dm², respectively.

In conclusion, the results confirm that the tested material complies fully with European food contact safety requirements, showing minimal overall migration and suitability for a broad range of food types, including aqueous, acidic, alcoholic, and fatty matrices.

Edible ink printing

The starch film with and without Broccoli extract were printed by edible inks through Canon printer (PIXMA) model iX6840.

As shown in Fig. 4, the visual inspection of the prepared films clearly demonstrates that all samples formed continuous, free-standing films with acceptable integrity, indicating good film-forming ability of starch as a bio polymeric matrix. The control film, blank sample, (S1), composed solely of starch, exhibited a relatively smooth, homogeneous, and translucent appearance, which is characteristic of gelatinized starch films due to uniform polymer chain dispersion and hydrogen bonding.

Incorporation of broccoli extract at increasing concentrations (S2–S4) led to gradual changes in film color, opacity, and surface texture. Films containing 1% broccoli extract (S2) maintained acceptable transparency with a slightly yellowish hue, while higher concentrations (3% and 5% in S3 and S4, respectively) resulted in more pronounced coloration and increased turbidity. These changes could be related to the existence of phenolic compounds, chlorophyll derivatives, in addition to other bioactive constituents in broccoli extract, which interact with the starch matrix and scatter incident light. Similar observations have been reported for starch films enriched with plant extracts and natural antioxidants [32, 33].

Despite increased extract loading, all films preserved structural continuity without visible cracks, indicating that broccoli extract did not compromise the cohesion of the polymer network. This suggests favorable compatibility between starch and the phytochemical constituents of broccoli.

The addition of broccoli extract is expected to enhance intermolecular interactions within the starch matrix.

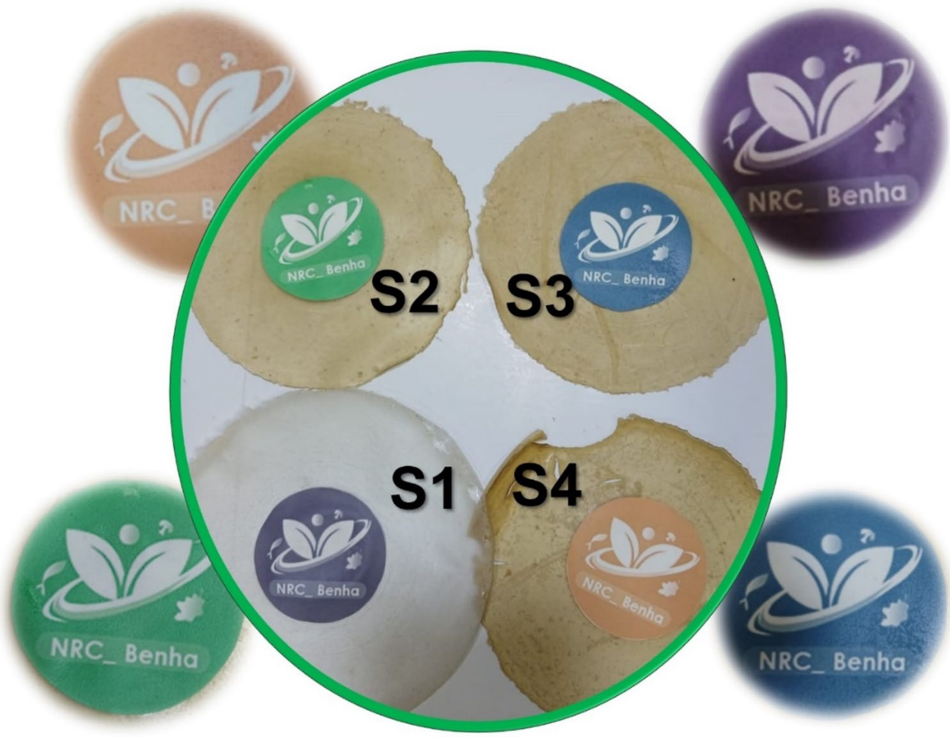


Fig. 4 Soluble starch films with different ratios of broccoli extract as substrate for printed edible labels

Hydroxyl groups from starch chains can form hydrogen bonds with phenolic compounds, flavonoids, and organic acids present in broccoli extract. At low concentrations (S2), these interactions may improve polymer chain packing and interfacial adhesion, resulting in uniform films [34].

Films S2 and S3 are anticipated to exhibit optimal barrier performance due to balanced hydrophilic/hydrophobic interactions, while S4 may show increased water vapor permeability due to excessive hydrophilic compounds disrupting the polymer network. Similar trends have been reported for starch-based films containing vegetable and fruit extracts [35, 36].

All films successfully supported printed images using edible inks, demonstrating good surface wettability and ink adhesion. The starch-based matrix provides a porous yet cohesive surface that enables ink absorption without bleeding or significant diffusion. The presence of broccoli extract did not negatively affect print clarity; instead, films S2 and S3 exhibited improved ink contrast, likely due to reduced surface gloss and enhanced ink anchoring.

At higher extract concentration (S4), slight surface irregularities may influence ink uniformity; however, image integrity remained acceptable. These findings confirm the suitability of starch/broccoli films for printed active packaging, which is essential for branding, labeling, and consumer communication UX (User Experience) UI (User Interface).

The progressive enrichment of films with broccoli extract (S2–S4) is expected to significantly enhance antioxidant activity, enabling these materials to function as active packaging systems. Antioxidants released from the film matrix can delay lipid oxidation, reduce microbial growth, and extend food shelf life. Film S3 (3% broccoli extract) is likely to represent the optimal formulation, balancing antioxidant efficiency, mechanical integrity, and printability. This active functionality in accordance with current trends in sustainable food packaging, whereas materials are designed not only as passive barriers but also as contributors to food preservation and safety [37].

All components used in the developed films, starch, broccoli extract, and edible inks, are biodegradable, renewable, and food-grade, increase their suitability for edible and compostable packaging applications. The absence of synthetic polymers, toxic solvents, or heavy metals enhances consumer safety and environmental compatibility. These films are particularly suitable for fresh produce packaging, dry food wrapping, single-use edible labels and sachets, and functional food packaging with health-promoting benefits.

The results clearly demonstrate that starch films incorporated with broccoli extract and printed with edible inks represent a promising class of eco-friendly, active, and printable packaging materials. Among the tested formulations, S2 and S3 offer the best balance between functionality, structural integrity, antioxidant activity, and print quality. The developed system addresses key challenges in biodegradable packaging and in accordance with worldwide efforts to decrease plastic waste while improving food safety and quality. Consistent with SEM images and printable starch film, there is agreement between the surface roughness with the ratio of broccoli and the quality of edible ink print processing [38].

Conclusion

Cationic potato-peel starch/broccoli, low cost and eco-friendly biodegradable packaging film, was successfully prepared by cast method. Blending of potato-peel starch enhanced the flexibility and ductility of the prepared film. It also improves resistance of the film to moisture and gas transmission. SEM pictures reveal that introducing broccoli to cationic potato-peel starch increases surface roughness and porosity which enhanced its printability susceptibility. DSC results reflect a reduction in crystallinity by the incorporation of broccoli extract, which is of importance to film wettability and ink adhesion. The overall migration results for all food simulants used were below the specified limit of 10 mg/dm^2 specified by the EU Commission Regulation (EU) 10/2011. This confirm that the tested material complies fully with European food contact safety requirements. In conclusion, Cationic potato-peel starch/broccoli films were successfully printed with edible inks and represent a promising class of environmental, active, and printable packaging materials. These biodegradable printable packaging films valorizing agro-waste, reducing plastic waste, and improving food safety and quality to great extent.

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Declarations

Conflict of interest The authors have no competing interests to declare that are relevant to the content of this manuscript.

References

1. P. Ayassamy, Ocean plastic pollution: A human and biodiversity loop. Environ. Geochem. Health. **47**, 1–17 (2025)
2. B.P.S. Gautam, A. Qureshi, A. Gwasikoti, V. Kumar, M. Gondwal, Global scenario of plastic production, consumption, and waste generation and their impacts on environment and human health. In Adv. Strat. Biodegradation of Plast. Polym. 1–34, (2024)
3. A. Maurya, P. Kumar, B. Kumar, V.K. Singh, Plastic waste: A long-lasting threat to the environment, in Global Waste Management. Chap. 13, pp. 334–358 (2025)
4. L. Petrenko, S. Puzko, V. Lavrenenko, I. Gernego, Fostering sustainable packaging industry: Global trends and challenges. Eur. J. Sustain. Dev. **13**(2), 63 (2024)
5. S.K. Verma, A. Prasad, V. Sonica, Katiyar, State of art review on sustainable biodegradable polymers with a market overview for sustainability packaging. Mater. Today Sustain. **26**, 100776 (2024)
6. E. Agama-Acevedo, P.C. Flores-Silva, L.A. Bello-Perez, Cereal starch production for food applications. In Starches for Food Application: Chemical, Technological and Health Properties, 71–102 (2019)
7. B.L. Tagliapietra, M.H.F. Felisberto, E.A. Sanches, P.H. Campelo, M.T.P.S. Clerici, Non-conventional starch sources. Curr. Opin. Food Sci. **39**, 93–102 (2021)
8. E.M. Nagy, C. Cota, N. Cioica, Z. Gyorgy, M. Todica, O. Cozar, FT-IR investigation of starch-based composite reinforced with Miscanthus fibers. AIP Conf. Proc. **1917**, 040009 (2017)
9. B. Iamareerat, M. Singh, M.B. Sadiq, A.K. Anal, Reinforced cassava starch based edible film incorporated with essential oil and sodium bentonite nanoclay as food packaging material. J. Food Sci. Technol. 55, 1953 (2018)
10. J. Bemiller, R. Whistler, *Starch: Chemistry and Technology* (Academic Press, Inc., Cambridge, MA, USA, 2009), pp.374–564
11. Y. Liu, M. Liu, L. Zhang, W. Cao, H. Wang, G. Chen, S. Wang, Preparation and properties of biodegradable films made of cationic potato-peel starch and loaded with curcumin. Food Hydrocoll. **130**, 107690 (2022)
12. M. Hasan, R. Rusman, I. Khaldun, L. Ardana, M. Mudatsir, H. Fansuri, Active edible sugar palm starch-chitosan films carrying extra virgin olive oil: Barrier, thermo-mechanical, antioxidant, and antimicrobial properties. Int. J. Biol. Macromol. **163**, 766–775 (2020)
13. V.D. Medeiros Silva, M.C. Coutinho Macedo, C.G. Rodrigues, A. Fante, Biodegradable edible films of ripe banana peel and starch enriched with extract of Eriobotrya japonica leaves. Food Bioscience. **38**, 100750 (2020)
14. N. Bhati, P. Bhatt, V. Kumar, S. Tomar, V.A. Sethi, A. Mittal, L.P. Maguluri, R. Goel, L.K. Tyagi, Eco-friendly enzymatic starch modification: precision strategies for tailored industrial and biomedical functionalities. Starch **77**(9), 70087 (2025)
15. R.K. Gupta, U.G.M. Eswaran, S. Kumar, S. Pipliya, B. Kaur, P.P. Srivastav, Synergistic Modification of Starch Polysaccharide via Sequential Treatments With Ultrasonic and Cold Plasma Technology. J. Food Process. Preserv. **185**, 1–17 (2025)
16. M.S. Gularte, R.F.N. Quadrado, N.S. Pedra, M.S.P. Soares, N.P. Bona, R.M. Spanevello, A.R. Fajardo, Preparation, characterization and antitumor activity of a cationic starch-derivative membrane embedded with a β-cyclodextrin/curcumin inclusion complex. Int. J. Biol. Macromol. **148**, 140–152 (2020)
17. A. Matloob, H. Ayub, M. Mohsin, S. Ambreen, F.A. Khan, S. Oranab, A review on edible coatings and films: advances, composition, production methods, and safety concerns. ACS Omega. **8**, 28932–28944 (2023)
18. L. Hakim, R.K. Deshmukh, Y.S. Lee, K.K. Gaikwad, Edible ink for food printing and packaging applications. Sustain. Food Technol. **2**, 876–892 (2024)
19. S. Dave, J. Das, B. Varshney, V.P. Sharma, Dyes and pigments: interventions and how safe and sustainable are colors of life, trends and contemporary technologies for photocatalytic degradation of dyes. 1–20 (2022)
20. -L.L.D.R. Osorio, E. Florez-Lopez, C.D. Grande-Tovar, The potential of selected agri-food loss and waste to contribute to a circular economy: applications in the food, cosmetic and pharmaceutical industries. Molecules. **26**, 515 (2021)
21. -I. Chiocchio, M. Mandrone, P. Tomasi, L. Marincich, F. Poli, Plant secondary metabolites: an opportunity for circular economy. Molecules. **26**, 495 (2021)
22. C.M.C. Andres, J.M. Perez de la Lastra, E.B. Munguira, C.A. Juan, E. Perez-Lebena, The multifaceted health benefits of broccoli-a review of glucosinolates, phenolics and antimicrobial peptides. Molecules. **30**(11), 2262 (2025)
23. M.D. Torres, P. Fradinho, P. Rodriguez, E. Falque, V. Santos, H. Dominguez, Biorefinery concept for discarded potatoes:

recovery of starch and bioactive compounds. J. Food Eng. **275**, 109886 (2020)
24. C.M.R. Almeida, J.M.C.S. Magalhaes, H.K.S. Souza, M.P. Goncalves, The role of choline chloride-based deep eutectic solvent and curcumin on chitosan films properties. Food Hydrocoll. **81**, 456–466 (2018)
25. -S. Ibrahim, H. Fahmy, S. Salah, Application of interactive and intelligent packaging for fresh fish shelf-life monitoring. Front. Nutr. **8**, 677884 (2021)
26. S.S. Ragab, M. Abdelraof, A.A. Elrashedy, A.M.K. Sweed, Design, synthesis, molecular dynamic simulation studies and antibacterial evaluation of new spirocyclic aminopyrimidines. J. Mol. Struct. **1278**, 134912 (2023)
27. M. El-Bendary, M. Abdelraof, M. Moharam, E. Elmahdy, M.A. Allam, Potential of silver nanoparticles synthesized using low active mosquitocidal *Lysinibacillus sphaericus* as novel antimicrobial agents. Prep. Biochem. Biotechnol. **51**(9), 926–935 (2021)
28. S. Sakat, A.R. Juvekar, M.N. Gambhire, In vitro antioxidant and anti-inflammatory activity of methanol extract of *Oxalis corniculata* Linn. Int. J. Pharm. Pharm. Sci. **2**(1), 146–155 (2010)
29. V. Kola, I.S. Carvalho, Plant extracts as additives in biodegradable films and coatings in active food packaging. Food Biosci. **54**, 102860 (2023)
30. E. Apostolidis, D. Kioupis, G. Kakali, N.G. Stoforos, I. Mandala, Effect of starch concentration and resistant starch filler addition on the physical properties of starch hydrogels. J. Food Sci. **86**(12), 5340–5352 (2021)
31. R.K. Deshmukh, K.K. Gaikwad, Natural Antimicrobial and Antioxidant Compounds for Active Food Packaging Applications. Biomass Convers. Biorefin. **14**, 4419–4440 (2024)
32. Y. Zhang, L. Ziheng, J. Zeng, H. Gao, J. Qi, Barrier properties characterization and release kinetics study of citrus fibers-reinforced functional starch composites. Food Hydrocoll. **164**, 111195 (2025)
33. S.A. Mir, B.N. Dar, A.A. Wani, M.A. Shah, Effect of plant extracts on the techno-functional properties of biodegradable packaging films. Trends Food Sci. Technol. **80**, 141–154 (2018)
34. D. Liu, P. Zhao, J. Chen, Y. Yan, Z. Wu, Recent Advances and Applications in Starch for Intelligent Active Food Packaging. Rev. Foods. **11**(18), 2879 (2022)
35. J. Bonilla, E. Talon, L. Atares, M. Vargas, A. Chiralt, Effect of the incorporation of antioxidants on physicochemical and antioxidant properties of wheat starch-chitosan films. J. Food Eng. **118**, 271–278 (2013)
36. M. Sapper, A. Chiralt, Starch-based coatings for preservation of fruits and vegetables. Food Chem. **8**(5), 152 (2018)
37. S. Yildirim, B. Rocker, M.K. Pettersen, J. Nilsen-Nygaard, Z. Ayhan, R. Rutkaite, T. Radusin, P. Suminska, B. Marcos, V. Coma, Active Packaging Applications for Food, Compr. Rev. Food Sci. Food Saf. **17**, 165–199 (2018)
38. Z. Domzalska, E. Jakubczyk, Characteristics of Food Printing Inks and Their Impact on Selected Product Properties. Foods. **14**(3), 393 (2025)