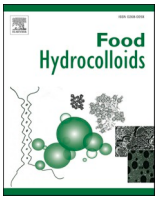
Food Hydrocolloids 133 (2022) 107888

Contents lists available at ScienceDirect

Food Hydrocolloids

journal homepage: www.elsevier.com/locate/foodhyd



Feasibility of active biobased films produced using red chilto wastes to improve the protection of fresh salmon fillets via a circular economy approach

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ARTICLE INFO

Keywords:
Active films
Valorization
Fish preservation
Polyphenol extracts
Pectin
Solanum betaceum

ABSTRACT

By-products of a native fruit of the Argentina northwest have been valorized for the extraction of both pectin and bioactive extracts with the aim of developing antioxidant films with potential application as food packaging materials. Initially, the composition and antioxidant properties of polyphenol-rich, anthocyanin-rich, and pectin extracts obtained from the seed and peel of *Solanum betaceum* (chilto) red fruits were characterized. Based on their higher antioxidant properties, peel extracts were selected as active compounds and incorporated in a film-forming matrix based on the pectin extract. Dry films were evaluated concerning their morphological, optical, thermal, mechanical, and water barrier properties. The developed antioxidant films were directly applied on salmon fillets, effectively improving their shelf-life and reducing lipid and protein oxidation during 10 days of storage at 4 °C. It was also found that the films containing peel-polyphenolic extract were the most promising, in agreement with their better barrier properties.

1. Introduction

The high consumers' demand for safe and healthier food, together with the increased awareness of the negative environmental impact of non-biodegradable packaging, has encouraged scientists to develop novel alternatives to plastic packaging which can help increasing products shelf-life through the use of natural compounds. In fact, there is a growing interest in the food industry to develop sustainable packaging materials (i.e., biodegradable coatings and films) to improve the food quality and shelf-life and to prevent food waste. However, biobased plastics do not fulfill the high-performance standards required to protect food products against contamination or the loss of food quality. Moreover, most of the starting raw materials are vegetable resources whose primary use is as food sources. Therefore, there is an increasing need to implement new strategies for agricultural waste management in a circular biobased economy for the extraction of biopolymers. The development of biodegradable films based on agro-industrial plant products and by-products is an excellent opportunity to add value to these residues and reduce waste accumulation leading to positive impacts on the environment (Zhong, Godwin, Jin, & Xiao, 2020). Furthermore, agro-industrial waste is a source of bioactive compounds (i.e., polyphenols) that can serve as functional or active compounds in the biodegradable food package. The development of biobased films derived from biological resources and food waste offers a great opportunity since their biodegradability and environmental compatibility are ensured.

Biobased packaging materials with active properties can prolong shelf-life or enhance safety, improving the stability of oxidation-sensitive foodstuff or retarding the microbial growth in food products while maintaining or even enhancing the quality of the product (Falcó,

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https://doi.org/10.1016/j.foodhyd.2022.107888
Received 27 April 2022; Received in revised form 2 June 2022; Accepted 16 June 2022
Available online 22 June 2022

Randazzo, Sánchez, López-Rubio, & Fabra, 2019; Moreno et al., 2020; Nilsen-Nygaard et al., 2021; Pérez et al., 2021). The entrappment of certain active compounds in the biopolymer matrix, allowing them to interact with the food and/or environment, offers innovative solutions to maintain or prolong the useful life and improve food quality and safety (Almasi, Jahanbakhsh Oskouie, & Saleh, 2020). In this sense, active biobased films can interact with packaged food promoting improvements in sensory and nutritional attributes.

Solanum betaceum also known as "tree tomato", "chilto", or "tamarillo", is a tree species currently used for human consumption in a large region of Northwestern Argentina and, more recently, the sustainable crop of chilto is being carried out on a larger scale. Chilto cultivation extends from South America to subtropical areas such as New Zealand, southern Europe, and tropical areas of other continents such as India, Nepal and Southeast Asia. This fruit is widely cultivated in most of South America, but is used primarily for local consumption and often in the processed form. In Ecuador, until 2009 there were 3000 ha producing 25,000 tons for domestic consumption at a rate of approximately 1.5 kg/capita/year (Ojeda, Bermeo, Bastidas, & Muñoz, 2009). In Colombia, about 120,000 tons of chilto are produced per year on 6500 ha, of which the majority is consumed locally (Márquez, Otero, & Cortés, 2007). According to the Ministry of Agriculture and Livestock of Ecuador (MAG, 2018), the "tree tomato" ranks tenth among inter-Andean fruit crops in terms of yield, and fifteenth in terms of cultivated area. The cultivated and harvested areas, as well as the yield per hectare, show a growth trend in recent years. New Zealand produces 100 tonnes of chilto on about 100 ha, worth US$2.4 million in domestic sales and US $100000 in export sales from this country. The United States, Australia, Hong Kong, Singapore, Japan and the Pacific Islands are the main export markets for *S. betaceum* fruits (Diep, Rush, & Yoo, 2020). Chilto is usually processed, generating food waste (peel and seeds). The pulp, seed and peel represent 51.5, 39.4 and 9.1% of the total weight of the fresh fruit, respectively. Therefore, approximately half of the weight of the fruit (49%) is considered waste material. These wastes are a source of natural antioxidants such as phenolic compounds, anthocyanins, and carotenoids (Orqueda et al., 2017, 2020, 2021; Isla et al., 2020; Ojeda et al., 2009; Ordóñez, Cardozo, Zampini, & Isla, 2010). Previous reports studied the composition and functionality of pectin polysaccharides extracted from orange variety chilto (do Nascimento et al., 2015; Gannasin, Mustafa, Adzahan, & Muhammad, 2015; do Nascimento, Iacomini, & Cordeiro, 2016) and red variety chilto (Orqueda et al., 2022). In addition, the excellent functionality of polyphenolic ethanol extracts obtained from chilto seed, pulp, and peel powders have demonstrated antioxidant, hypoglycemic, and hypolipemic activity attributed to phenolic acids in their composition, such as rosmarinic and caffeoylquinic acid derivatives (Ordóñez et al., 2010; Orqueda et al., 2017, 2020, 2021). Zein fibers loaded with phenolic-enriched extracts from pulp, seed, and peel of orange chilto from Argentina were spun-coated on polyhydroxyalkanoate films, for their potential use as internal coatings with antioxidant properties contributing to the preservation of both hydrophilic and lipophilic food products (Moreno, Orqueda, Gómez-Mascaraque, Isla, & Lopez Rubio, 2019). Other *S. betaceum*-derived products are carotenoid-rich microencapsulates derived from the pulp of yellow chilto from Colombia (Garcia, Giuffrida, Dugo, Mondello, & Osorio, 2018).

To the best of our knowledge, there is no existing literature on the potential of polysaccharides, polyphenolic compounds, and anthocyanins from red chilto variety waste or by-products as bio-degradable edible films. Therefore, the main goal of this work was to carry out a potential valorization of red chilto sub-products (peel and seed) for developing active edible films. First, the chemical composition and antioxidant activity of polyphenolic and anthocyanin enriched extracts from seeds and peels of red chilto were compared. Also, the film-forming properties of pectin-enriched extracts from red chilto peel were evaluated. Finally, the effectiveness of the obtained films for reducing the oxidation of salmon fillets during storage was analyzed.

2. Materials and methods

2.1. Reagents

Diammonium salt of 2,2′ -azino-bis (3-etylbenzothiazolin-6-sulfonic) acid (ABTS), potassium persulphate ($K_2O_8S_2$), β-carotene, potassium iodide (KI), trichloroacetic acid (TCA), thiobarbituric acid (TBA), and Ellman's reagent 5,5′ -dithiobis (2- nitribenzoic acid) (DTNB) were purchased from Sigma-Aldrich. Ethanol 96% and sodium hydroxide (NaOH) were obtained from Scharlab.

2.2. Extraction and characterization of pectin and bioactive compounds of red chilto fruits

The fruits of *Solanum betaceum* (red variety) were harvested at the ripening stage for consumption in Finca del Obispo (Villa Jardín de Reyes, Jujuy, Argentina) in December 2016. Voucher specimens, rejected for commercialization due to minor damages in the peel or unacceptabvle appearance, were deposited at the Fanerogamic Herbarium of Fundación Miguel Lillo (LIL-HbF), Tucuman, Argentina (Voucher number 617.907/LIL). Peel, pulp, and seeds were separated, freeze-dried, and powdered. The yield in grams of powder per 100 g of fresh fruit was 26.0, 20.4 and 25.9% for peel, pulp and seed, respectively. The powders obtained were vacuum-packed and stored at −20 °C.

Polyphenols and anthocyanins-enriched extracts were obtained from the red chilto seed and peel powders. The polyphenol extracts were obtained according to the methodology described by Orqueda et al. (2020). Briefly, seed and peel powders were extracted with a 95% (v/v) ethanol solution (1:5 w/v solid:liquid ratio) at room temperature and applying an ultrasound treatment for 30 min. The anthocyanin extraction methodology was similar to that used for polyphenols, but acidified ethanol (1% citric acid) was used as a solvent. Then, the obtained extracts were filtered and dried under reduced pressure (0.133 Pa and −45 °C) to produce the polyphenolic-enriched extracts (named as PoE-P and PoE-S depending on the extracting material, peel, and seeds, respectively) or anthocyanins-enriched extracts (named as AnE-P and AnE-S, for those obtained from peel and seed, respectively).

On the other hand, a pectin-enriched fraction (PeE-P) was obtained from red chilto peel, according to the methodology described by Orqueda et al., 2022. Briefly, 10 g of red chilto peel powder was dispersed in 300 mL of distilled hot water for 2 h at 100 °C. Then, the mixture was centrifuged at 1500 g for 20 min. The supernatants were combined, and the polysaccharides were precipitated with absolute ethanol (2 vol) and were recovered by centrifugation at 3000*g* for 10 min. The resulting polysaccharide was freeze-dried to obtain the pectin enriched extract (PeE-P).

2.2.1. Protein content

The protein content in the pectin, anthocyanins, and polyphenols enriched extracts was calculated from the nitrogen content with the Kjeldahl method (BÜCHI K350, Switzerland), multiplied by a factor of 6.25 (AACC 08–01, 2000). All determinations were performed in triplicate.

2.2.2. Carbohydrate composition

The monosaccharide composition was confirmed by acid hydrolysis, followed by chromatographic analysis (Morais de Carvalho et al., 2017). The samples were hydrolyzed with 2 mol L^{-1} trifluoroacetic acid (TFA) at 120 °C for 3 h. The samples were dried under a stream of air to evaporatoe the volatile solvent and dissolved in milliQ water. The monosaccharides were analyzed using high-performance anion-exchange chromatography with pulsed amperometric detection (HPAEC-PAD) using an ICS-6000 system (Dionex) equipped with a CarboPac PA1 column (4 × 250 mm, Dionex).

2.2.3. Methoxylation degree of pectin extracts

The degree of methoxylation (MD) of PeE-P was analyzed by the alcohol oxidase enzyme method, using methanol as a standard (Grassino et al., 2016). The PeE-P was treated with 1 N potassium hydroxide for 30 min at room temperature. Then, the mixture was neutralized using phosphoric acid. The hydrolyzed pectin was mixed with 1 mL of alcohol oxidase (1 U/mL) and incubated at 25 °C for 15 min. Then, 2 mL of 0.02 M 2,4-pentanedione was added and mixed vigorously. The reaction mixture was incubated at 60 °C for 15 min, and the absorbance was measured at 412 nm.

2.2.4. Ash content

The ash content in the extracts was determined by calcination and gravimetric analysis (AACC 08–01, 2000). The samples were weighed in a crucible and incinerated in a muffle at 550 °C for 5 h. Then, the ash content was calculated by weight difference.

2.2.5. Total anthocyanin and phenolic content

The content of total phenolic compounds in all extracts was measured using the Folin-Cicoalteu assay (Singleton, Orthofer, & Lamuela-Raventos, 1999). Different sample concentrations were mixed with 1000 μL of Folin-Ciocalteau reagent. Then, 800 μL of sodium bicarbonate (75 g L^{-1}) was added to the reaction mixture, and after incubation at 40 °C for 30 min, the absorbance at 760 nm was measured. The content of total phenolic compounds was calculated using a calibration curve with gallic acid, and the results were expressed as mg gallic acid equivalents in 100 g of the dry weight of extract (mg GAE/100 g DW). Total anthocyanins content was determined using the differential pH method described by Costamagna et al. (2013), and the results were expressed in mg of cyanidin-3-glucoside equivalents per 100 g of DW (mg C-3GE/100 g DW).

2.2.6. Total antioxidant activity of red chilto extracts

The total antioxidant activity of pectin, anthocyanins, and polyphenolic extracts was tested using the ABTS (2,2-azinobis (3-4ethylbenzothiazoline)-6-sulfonic acid) radical-cation method ($ABTS^{\bullet+}$) (Orqueda et al., 2017). A 7 mM ABTS solution was prepared in sodium phosphate buffer at pH 7.4 and mixed with 2.45 mM potassium persulphate. This solution was left overnight in the dark to yield the $ABTS^{\bullet+}$ radical cation. Before use, the $ABTS^{\bullet+}$ was diluted in sodium phosphate buffer, pH 7.4, with an initial absorbance of ~0.700 ± 0.02 at 734 nm, at room temperature. For the reaction mixture, 100 μL of each extract at different concentrations are added to 200 μL of $ABTS^{\bullet+}$ solution and mixed by shaking. Results were expressed as the extract concentration necessary to scavenge 50% of ABTS radical catión (SC_{50}).

2.2.7. β-Carotene-linoleic acid bleaching assay

The antioxidant activity of red chilto extracts was measured according to the β-carotene bleaching assay described by Fontes-Candia, Erboz, Martínez-Abad, López-Rubio, and Martínez-Sanz (2019). In brief, 2 mL of β-carotene solution (4 mg of β-carotene in 20 mL chloroform) was placed on a rotary evaporator, and the chloroform was evaporated. Then, 50 μL of linoleic acid and 400 mg of Tween 40 were added and stirred. After that, 100 mL of aerated distilled water were transferred to the flask and shaken vigorously to form a stable emulsion. 10 μL of sample (0.5–10 mg/mL in PBS) were added to 250 μL of the β-carotene-linoleic acid emulsion and incubated in the dark at 45 °C for 120 min. The absorbance was measured at 0 min and 120 min at 470 nm using a microplate spectrophotometer CLARIOstar (BMG LABTECH, Germany). PBS and *tert*-butyl hydroxytoluene (BHT) were used as the blank and positive control, respectively. The antioxidant activity (AA) of each extract was calculated.

2.3. Preparation and characterization of chilto-based films

Antioxidant films were prepared using the pectin-enriched extract (PeE-P) as a film-forming matrix and the red chilto peel polyphenolic enriched extract and anthocyanin enriched extracts (PoE-P and AnE-P) as active compounds. These extracts were selected based on their antioxidant activity and yield. Specifically, pectin films were produced by adding 1 g of pectin extract to 100 mL of distilled water and dissolving them under magnetic stirring for 30 min at a controlled temperature of 45 °C. Films containing anthocyanins enriched extract or polyphenolic extract were prepared by dispersing 0.5 g of AnE-P or PoE-P in the prepared pectin solution.

Afterward, film-forming dispersions were spread evenly over a Teflon casting plate (15 cm diameter) resting on a level surface. Films were formed by drying for approximately 24 h at 30 °C. Dry films could be peeled intact from the casting surface.

The obtained films were removed from the plates and equilibrated for four days in a desiccator at 20 °C and 53% relative humidity (RH) using an oversaturated solution of $Mg(NO_3)_2$. Film thickness was measured using a hand-held digital micrometer (Palmer-Comecta, Spain, 0.001 mm) at five random positions, and the average value was used in mechanical and water vapor permeability calculations.

2.3.1. Characterization of film morphology

The microstructure of the films was analyzed using a scanning electron microscope (Hitachi S-4800). Each film (in duplicate) was dipped in liquid nitrogen and then randomly broken to evaluate the cross-section. Films were mounted on M4 Aluminium Specimen Mount and fixed on the support using double-side adhesive tape. Samples were gold–palladium-coated and observed using an accelerating voltage of 10 kV.

2.3.2. Optical properties of bioactive films

The transparency of the films was determined through the surface reflectance spectra in a spectrocolorimeter CM-3600 d (Minolta Co., Tokyo, Japan), with a 10 mm^2 illuminated sample area. Measurements were taken in duplicate for each sample using white and black backgrounds.

Film transparency was evaluated through the internal transmittance (Ti) (0–1, theoretical range) by applying the Kubelka-Munk theory for multiple scattering to the reflection data. Moreover, CIE-L_{ab}* h_{ab}* Cab* coordinates were obtained from the reflectance of an infinitely thick layer of material.

2.3.3. Mechanical properties

Mecmesin MultiTest 1-i (1 kN) machine (Virginia, USA) with the Emperor™ software was used to determine tensile strength (TS), elastic modulus (E), and elongation (EAB) properties, according to ASTM standard method D882-09 18 (ASTM, 2010a). E, TS, and EAB properties were obtained from the stress-strain curves, estimated from force-distance data obtained for the different films. At least eight replicates of each film formulation were tested. Equilibrated specimens (1 cm wide and 8 cm long) were mounted in the film extension grips and stretched at 50 mm min^{-1} until breaking.

2.3.4. Fourier transform infrared spectroscopy (FT-IR)

Films were analyzed by FT-IR in attenuated total reflectance (ATR) mode using Thermo Nicolet Nexus (GMI, USA) equipment. The spectra were taken at 4 cm^{-1} resolution in a wavelength range between 800 and 4000 cm^{-1} and averaged a minimum of 32 scans.

2.3.5. Water vapor permeability (WVP)

Direct permeability to water was determined from the slope of the weight versus time curves at 25 °C and 53–100% RH gradient, according to the ASTM E96/E96M-10 (ASTM 2010b) gravimetric method. The films were sandwiched between the aluminum top (O-ring open) and the bottom (deposit for the distilled water) parts of a specifically designed payne permeability cups of 3.5 cm in diameter (Elcometer SPRL, Hermelle/s Argenteau, Belgium). The cups were placed in pre-equilibrated

Table 1
Chemical and biological characterization of pulp, seed and peel extracts from red chilto.

Samples	Composition				Antioxidant activity		
	Anthocyanins (mg C-3GE/100 g DW[a])	Total phenolics compounds (mg GAE/ 100 g DW[b])	Protein (%)	Ash (%)	Carbohydrates (%)	ABTS (SC_{50} μg/ mL[c])	β-carotene (% AA)[d]
AnE-S	320.6 ± 1.6[a]	481.8 ± 5.5[b]	2.9 ± 0.02[b]	1.6 ± 0.4[a]	23.4 ± 3.2	65.0 ± 1.4[c]	58.3 ± 1.3[c]
AnE-P	356.2 ± 2.3[b]	501.5 ± 2.1[c]	1.9 ± 0.2[a]	2.5 ± 0.1[b]	6.2 ± 1.0	14.4 ± 2.5[b]	61.5 ± 0.9[cd]
PoE-S	56.0 ± 0.6[c]	496.5 ± 3.0[c]	7.0 ± 0.6[d]	4.0 ± 0.4[c]	17.5 ± 0.5	12.9 ± 0.9[b]	52.9 ± 1.2[b]
PoE-P	68.2 ± 1.0[d]	705.6 ± 3.6[d]	5.7 ± 0.5[c]	7.0 ± 0.2[d]	10.3 ± 1.2	4.2 ± 0.6[a]	63.7 ± 1.6[d]
PeE-P	6.5 ± 0.5[e]	40.0 ± 2.0[a]	4.7 ± 0.2[c]	8.1 ± 0.1[e]	16.3 ± 2.7	510.0 ± 3.5[d]	35.4 ± 0.6[a]

AnE-S: Anthocyanin extract obtained from seeds; AnE-P: Anthocyanin extract obtained from peel; PoE-S: Polyphenols extract obtained from seed; PoE-P: Polyphenols extract obtained from peel. PeE-P: Pectin enriched extract. Mean values ± standard deviation. Values with different letters in the same column are significantly different ($p \leq 0.05$).

[a] mg of cyanidin 3-glucoside equivalents (C-3GE) per 100 g of dry weight (DW).

[b] mg of gallic acid equivalents (GAE) per 100 g of dry weight (DW).

[c] SC_{50}: Concentration of extract necessary to scavenge 50% of $ABTS^{\bullet+}$.

[d] At concentration of 20 μg/mL for AnE-S, AnE-P, PoE-S, and PoE-P, and 40 μg/mLfor PeE-P.

cabinets at 54% RH using magnesium nitrate saturated solution (Panreac Quimica, SA, Barcelona, Spain), and they were weighted periodically until the steady-state was reached. The free film surface during film formation (the air side) was exposed to the lowest relative humidity to simulate the actual application of the films in high water activity products when stored at intermediate relative humidity. The WVP tests were performed in quintuplicate.

2.4. Evaluation of the antioxidant effect of bioactive films on salmon fillets

Fresh salmon fillets from Atlantic salmon (*Salmo salar*) were purchased in a local market of Valencia (Spain). Fillets were cut into pieces (5 cm × 5 cm × 2 cm) and were randomly assigned into four groups: uncoated (control) and coated samples with the three different films (PeE-P, AnE-P, and PoE-P).

Fillets were coated on both sides with films prepared the day before and placed in the dark under refrigeration conditions (4 °C) for 8 days. Then, the quality of the stored salmon fillets was evaluated and compared regarding pH, lipid, and protein oxidation.

The pH values of salmon samples were determined following the protocol previously described by Xiong, Li, Warner, & Fang (2021) with slight modifications. Briefly, 2 g of each sample was homogenized with 20 mL of distilled water in an Ultraturrax homogenizer at 10,000 rpm for 40 s. The pH of the mixture was measured using a pH-meter at room temperature (25 °C). Each sample was measured in triplicate.

Lipid oxidation was evaluated using the thiobarbituric acid reactive substances (TBARS) assay according to the method described previously by Kulawik, Jamróz, Zając, Guzik, and Tkaczewska (2019). Briefly, 5 g of each sample of salmon were homogenized with 25 mL of 0.1% TCA and centrifuged at 10,000×*g* for 15 min 4 mL of the resultant supernatant were transferred in a test tube and mixed with 4 mL 20% TCA containing 0.67% of TBA. Then, 0.1 mL of BHT was added, and the mixture was heated at 90 °C for 30 min. Afterward, the reaction mixture was cooled at room temperature, and the absorbance was recorded at 532 nm using a spectrophotometer. The calculation was performed using the molar extinction coefficient for malondialdehyde of 155 mM^{-1} cm^{-1} and expressed as mg of TBARS/kg of salmon fillets.

The degree of lipid oxidation in the fish samples after 10 days of storage was also measured by following the peroxide index method (Pérez et al., 2021). In brief, 5 g of each sample was homogenized with 50 mL chloroform:methanol (2:1 v/v). Then, 10 mL of $CaCl_2$ solution were added to the mixture and shaken vigorously for 15 s. Afterward, the mixture was centrifuged at 4500×*g* for 30 min and evaporated under vacuum. After that, the extracted lipid content was then subjected to peroxide value (PV) analysis. One gram of salmon oil was placed in a flask and dissolved in a mixture containing chloroform and acetic acid (2:3 v/v) and an oversaturated KI solution (0.5 mL). Next, the flask was kept in the dark for 5 min, and 30 mL of distilled water were added. The PV was determined by titration of the mixture with a solution of sodium thiosulphate (0.001 N), using a starch solution (1%) as the indicator. A blank without sample was also titrated under the same condition. The peroxide value was expressed as peroxide milliequivalents per kilogram of lipid.

Finally, Ellman's reagent 5,5′ -dithiobis (2- nitribenzoic acid) (DTNB) was used to determine the protein oxidation by quantifying the free thiol group content in the salmon samples (Lund, Lametsch, Hviid, Jensen, & Skibsted, 2007). In brief, 2 g of fish sample was homogenized in 50 mL of 5% sodium dodecyl sulfate (SDS) in Tris buffer (100 mM, pH 8.0) at 12,000 rpm for 60 s. The resulting mixture was heated in a water bath at 80 °C for 30 min, and after cooling the sample, the protein content of the solution was determined by the Lowry reagent method (Peterson, 1977) using bovine serum albumin as the standard. For thiol group content determination, the protein concentration of the mixture was adjusted to 1.5 mg/mL by dilution with 5% SDS in Tris buffer (100 mM, pH 8.0). The 0.5 mL of the diluted filtrate was mixed with 2.0 mL Tris buffer (100 mM, pH 8.0) and 0.5 mL 10 mM DTNB in Tris buffer (100 mM, pH 8.0) and incubated for 30 min to develop the color. The absorbance of the mixture was measured using the spectrophotometer at 412 nm against a blank solution consisting of 0.5 mL of 5% SDS in Tris buffer (100 mM, pH 8.0) and 2.5 mL Tris buffer (100 mM, pH 8.0). L-cysteine was used as the standard. The level of free thiol groups in the *Salmon salar* fillets was calculated by dividing the thiol group content by the protein content, and results were expressed as μmol thiol/mg protein.

2.5. Statistical analysis

Each experimental value is expressed as the mean ± standard deviation. The statistical analysis of experimental data was performed using InfoStat software (Student Version, 2011). The one-way ANOVA with Tukey post-test at a confidence level of 95% was used to evaluate the significance of differences between groups. The criterion of statistical significance was taken as $p \leq 0.05$.

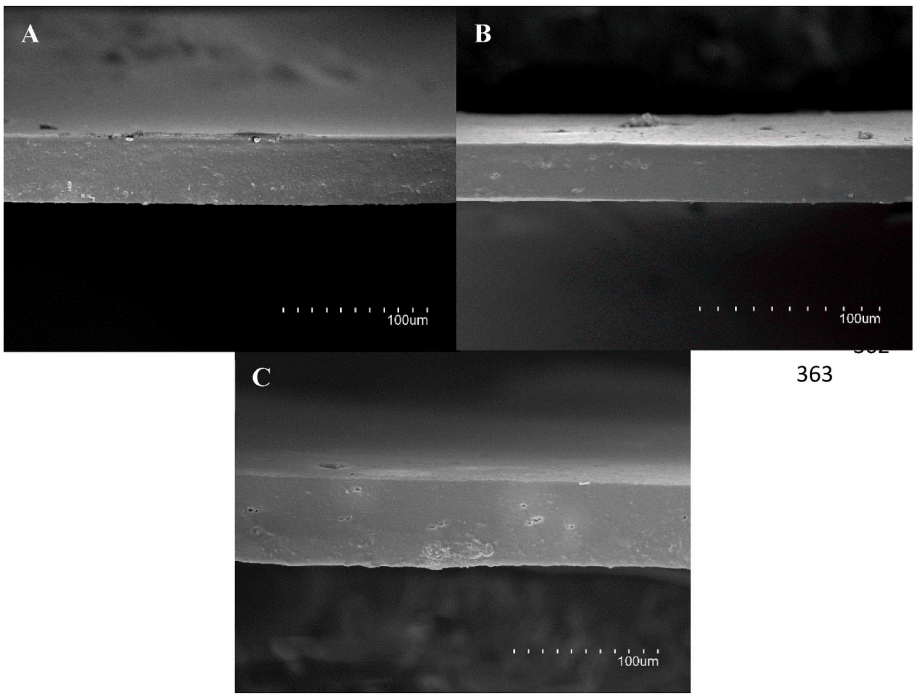

Fig. 1. Cross-section of films. A) Films of pectin from red chilto peel (PeE-P), B) Films containing anthocyanins extract from red chilto peel (AnE-P) and C) Films containing polyphenols extract from red chilto peel (PoE-P).

3. Results and discusion

3.1. Characterization of red chilto extracts

In the first part of this work, pectin and polyphenol-enriched extracts obtained from red chilto fruits were characterized to evaluate the possibility of generating antioxidant films for food applications. The results in Table 1 show the chemical and functional characterization of the extracts. The samples with the highest content of phenolic compounds were those obtained from chilto peel, both AnE-P and PoE-P. Furthermore, these peel extracts showed the highest response through the β-carotene bleaching assay, a practical test for evaluating antioxidant performance. Phenolics account for the main compounds in all extracts, except for the pectin extract, and slight variations in the protein, ash, and carbohydrate content were observed. Anthocyanin extracts showed relatively low contents in protein and also of ash impurities when compared with the polyphenol or pectin extracts (Table 1). The lower ash and protein content and higher ABTS antioxidant response pointed towards a higher amount of free phenolic compounds. This tendency was observed in AnE-P extracts compared to PoE-P extracts and in seed extracts compared to peel extracts. The carbohydrate composition in the anthocyanin and polyphenolic ethanolic extracts from seed and peel of red chilto consisted mainly of glucose, probably from free glucose or sucrose present in the fruit, with negligible quantities of rhamnose and galactose, while pectin-enriched extracts contained typical pectin components, mainly galacturonic acid, as well as arabinose, rhamnose and galactose (Table S1). The methoxylation degree of red chilto pectin was 60.8%, therefore it can be considered highly methoxylated pectin (HM-pectin; DM>50%). Based on the lower amount of impurities, higher phenolic content and excellent antioxidant performance, AnE-P and PoE-P extracts were selected, while the pectin-enriched extract was used as a film-forming matrix to develop antioxidant chilto films for food packaging applications.

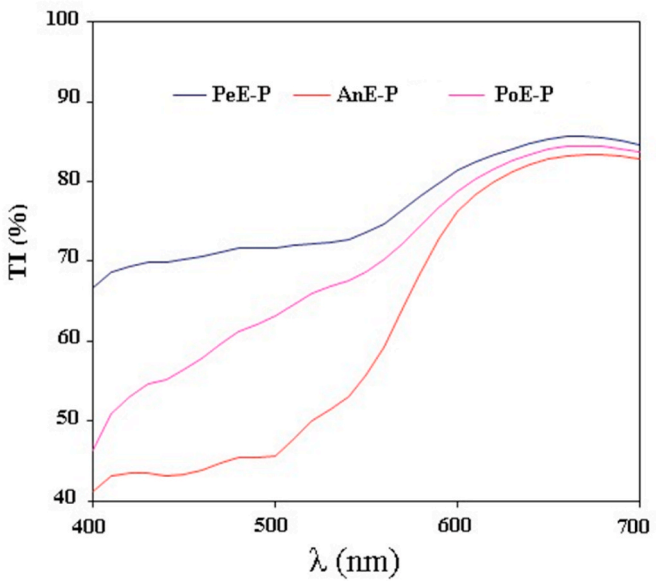

Fig. 2. Spectral distribution of internal transmittance (Ti) of the developed stand-alone films. Films of pectin of red chilto peel (PeE-P) and containing anthocyanins extract from red chilto peel (AnE-P) and polyphenols extract from red chilto peel (PoE-P). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

3.2. Characterization of bioactive films

3.2.1. Morphology

Fig. 1 shows the SEM micrographs of films pre-conditioned at 53% RH. The cross-section of the neat pectin extract revealed a homogeneous, continuous and smooth microstructure without cracks or bubbles, evidencing the formation of a well-ordered three-dimensional structure of pectin chains. The addition of both extracts provided a similar film structure where the AnE-P and PoE-P were entrapped in the

Table 2
Colour parameters of the developed stand-alone films.

	L*	C^*_{ab}	h^*_{ab}
PeE-P	**54.1**	**18.9**	**34.0**
AnE-P	**31.3**	**31.3**	**38.0**
PoE-P	**48.0**	**24.5**	**51.0**

L* lightness, C^*_{ab} Chroma, h^*_{ab} hue.
Films of pectin of red chilto peel (PeE-P) and containing anthocyanins extract from red chilto peel (AnE-P) and polyphenols extract from red chilto peel (PoE-P).

Table 3
Mechanical[a] and barrier[b] properties of the bioactive films.

	Thickness (mm)	E (MPa)[a]	TS (MPa)[a]	EAB (%)[a]	WVP 10^{10} (g-/Pa-*s*-m^2)[b]
PeE-P	0.108 ± 0.003[a]	1112 ± 242[b]	14.3 ± 3.7[a]	1.5 ± 0.4[a]	6.2 ± 0.1[ab]
AnE-P	0.112 ± 0.012[a]	409 ± 79[a]	11.6 ± 1.4[a]	5.4 ± 0.8[b]	6.7 ± 0.1[b]
PoE-P	0.110 ± 0.011[a]	500 ± 90[a]	9.5 ± 0.8[a]	2.7 ± 0.6[a]	5.4 ± 0.6[a]

Films of pectin of red chilto peel (PeE-P) and containing anthocyanins extract from red chilto peel (AeP-P) and polyphenols extract from red chilto peel (PoE-P).
[a] Elastic modulus (E), tensile strength (TS), and elongation at break (EAB).
[b] Water vapor permeability (WVP). Different superscripts within a column indicate significant differences among formulations ($p < 0.05$). Mean values ± standard deviation.

pectin matrix, showing a smooth and homogenous phase in which, some irregular PoE-P particles are embedded in the biopolymer matrix. It was more evident in the PeE-P-pectin films due to their lower water solubility than AnE-P. Similar results were reported by Shivangi, Dorairaj, Negi, and Shetty (2021), where the incorporation of ethanolic extracts of mulberry did not significantly affect the microstructure of the pectin films.

3.2.2. *Physicochemical characterization of pectin-based films*

Since these films were intended to be used in food packaging applications, optical properties (transparency and color parameters) are attributes of interest to be evaluated. It is well-known that the internal network of these films usually affects different properties, including their optical properties, given that changes in the refractive index occur through the polymer matrix. Fig. 2 plots the spectral distribution curves of Ti of films as a transparency indicator. High values of Ti are associated with more transparent films with a more homogeneous refractive index through their structures, whereas lower Ti values correspond to more opaque films with heterogeneous networks. As it was observed, all the films presented a similar pattern over the wavelength range considered. Films prepared with the pectin extract displayed higher transmittance values (i.e., high transparency). The addition of chilto extracts (PoE-P and AnE-P) significantly reduced the transparency of polysaccharide-based films, showing lower Ti values in all the wavelengths considered, indicating that these films were less transparent. A particular decrease in the Ti of the films between 400 and 550 nm was observed in all cases, even in the neat pectin films, evidencing a selective absorption of the red-pink components of the extracts (PoE-P and AnE-P) and confirming the presence of some polyphenols (as shown in Table 1) linked to the pectin extract since neat pectin films also showed a red-pink tonality. This decrease in the intensity of the Ti values was higher in the case of AnE-P films, ascribed to the native colour of anthocyanins which are very abundant in chilto peel (Orqueda et al., 2020). Lower Ti values in the UV–Vis spectra could be favourable for food protection by providing a light barrier for some food components easily degraded by light, such as lipids (Zhang, Jiang, Rhim, Cao, & Jiang, 2021). Table 2 summarizes the values of color coordinates (L*, lightness; C^*_{ab}, chrome; h^*_{ab}, hue) of the different films. As expected, films with PoE-P and AnE-P were darker (lower L*) with more saturated reddish colour for AnE-P films (higher C $^*_{ab}$ values) and brownish for PEE films (higher h^*_{ab}).

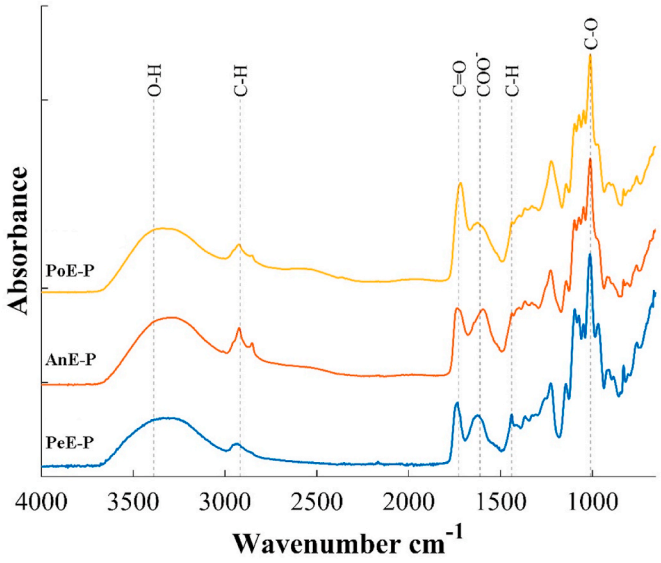

Fig. 3. FTIR spectra of films of pectin of red chilto peel (PeE-P) and containing anthocyanins extract from red chilto peel (AnE-P) and polyphenols extract from red chilto peel (PoE-P). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

The mechanical properties were assessed through tensile testing, and the most representative parameters are summarized in Table 3. When comparing the films with and without extracts, the first evident observation was that the presence of PoE-P and AnE-P had a significant impact on the mechanical properties. The neat pectin-films presented significantly higher elastic modulus and tensile strength and lower elongation at break than PoE-P and AnE-P. The higher elongation of pectin films containing chilto extracts can be ascribed to the potential interactions between hydrogen bonds of anthocyanins and the pectin side chains and also, to the presence of small sugars which could have a plasticizer effect on the biopolymer network. Specifically, the glycosidic nature of anthocyanins may explain the higher increase in the extensibility of the AnE-P film. Assis, Lopes, Costa, Flôres, and de Oliveira Rios (2017) noted that an increase in the disorder of the network of polymers would promote the formation of discontinuities in the structure, causing a lower value of TS. Giménez, De Lacey, Pérez-Santín, López-Caballero, and Montero (2013) also reported a decrease in TS due to polyphenolic compounds incorporation into polysaccharide-based films, ascribed to the reduction in the molecular interactions between polysaccharide side chains.

Barrier properties of the films are usually described by their permeabilities values. The results, compiled in Table 3, suggest no big differences between the samples. In general, WVP values ranged between 5.4 and 6.7 10^{-10} g Pa^{-1} s^{-1} m^{-2} in agreement with values previously reported for pineapple and orange pectin films (Rodsamran & Sothornvit, 2019, Spatafora Salazar, Sáenz Cavazos, Mújica Paz, & Valdez Fragoso, 2019). The incorporation of PoE-P extract slightly improved the barrier performance of the pectin films. It might be again related to the prevalence of less hydrophilic compounds in the ethanolic extract (PoE-P) in comparison to the AnE-P extract.

Infrared spectroscopy allowed investigating the potential interactions between the extracts and the neat pectin-based matrices. ATR-FTIR spectra related to pectin-based films containing or not PoE-P or AnE-P are gathered in Fig. 3. In general, all the spectra showed a broad absorption band in the 3600 - 3000 cm^{-1} spectral region, ascribed to the

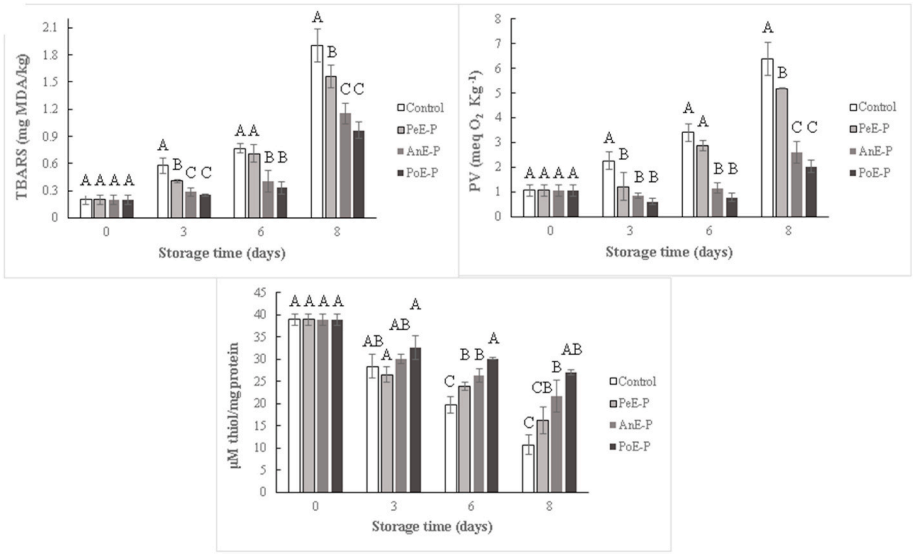

Fig. 4. Changes in TBARS values (mg MDA/kg meat) (**A**); Peroxide values (meq O_2 Kg^{-1}) (**B**) and free thiol group values (μmol thiol/mg protein) (**C**) of salmon fillet samples during cold storage (4 °C) for 8 days. Films of pectin of red chilto peel (PeE-P) and containing anthocyanins extract from red chilto peel (AnE-P) and polyphenols extract from red chilto peel (PoE-P).The value on day 0 was the initial value (control), and the bar was repeated on day 0 for clarity and comparison. Bars with different letters within the same day are significantly different at $P < 0.05$.

O–H stretching groups of the intra- and intermolecular hydrogen bonds of the galacturonic acid polymer (Gharibzahedi, Smith, & Guo, 2019). The bands located at 2918 and 2850 cm^{-1} region corresponded to the C–H stretching vibrations. In theory, the O–CH_3 stretch bands in the region 1600-1800 cm^{-1} could be used to differentiate low methoxyl pectins (LMP) and high methoxyl pectins (HMP). The strongest bands in the 1760-1745 cm^{-1} region were associated with the vibration of the esterified carbonyl group C═O, while the 1640-1620 cm^{-1} bands were due to the free carboxylic groups COO-. Both peaks are widely used to estimate the degree of esterification in pectins, which can be calculated as the ratio between the peak area of the band corresponding to the carboxylate esterified group and the sum of both mentioned bands (Kyomugasho et al., 2015). The carbonyl ester band is more intense in HMP than in LMP pectins, and therefore, according to the resulting FT-IR spectrum, the pectin extract is a HMP, confirming the results commented above. The band located at 1622 cm^{-1} slightly shifted to lower wavelengths in AnE-P films spectra suggesting the interaction between polyphenols and pectin. Pure pectin provides hydrophilic domains and hydrophobic pockets where polyphenols can be retained (Le Bourvellec, Guyot, & Renard, 2004). The pectin-anthocyanin interactions are mainly governed by hydrogen bonds formed by hydroxyl groups of the B ring of anthocyanin and dissociated galacturonic acid in the homogalacturonan chain (Larsen, Buerschaper, Schieber, & Weber, 2019). Furthermore, the acidic pH of the pectin solution enhances the interactions by facilitating the ionic interactions between the negatively charged galacturonic acid residues and the positively charged flavylium cations of anthocyanins. Thus, the FTIR characterization allowed to identify the groups involved in the interactions between films' components and confirmed the assumption from the mechanical behaviour of the films.

3.3. Evaluation of the antioxidant effect of the edible films on salmon fillets

As a final proof of concept, neat pectin films and those containing PoE-P or AnE-P were tested as packaging materials at refrigerated temperatures to inhibit lipid and protein oxidation during the storage of salmon fillets. To this end, salmon fillets were directly covered with the corresponding films and stored at refrigerated conditions during 8 days. The oxidative stability of salmon fillets was measured at 0, 3, 6, and 8 days of storage, simulating typical shelf-life and marketing conditions. The TBARS assay is widely used to evaluate lipid oxidation, and the results are displayed in Fig. 4a. The initial TBARS value was 0.22 mg MDA/kg of fish and increased in the different samples during the storage period, although this increase was significantly ($P < 0.05$) lower in the samples coated with the AnE-P and PoE-P films. Interestingly, after 8 days of storage, the salmon fillets coated with PoE-P films presented values of TBARS lower than 1 mg MDA/kg of fish, indicating that this film protected fresh salmon samples against lipid oxidation. It is worth mentioning that TBARS values higher than 2 mg MDA/kg are usually associated with a negative impact on the sensory attributes (Connell, 1995), a value that was not reached in any of the coated samples. The protective effect of antioxidant-containing films could be ascribed to the antioxidant activity of PoE-P and AnE-P compounds such as derivatives of rosmarinic acid and caffeoylquinic acid (Orqueda et al., 2020). In this context, the effective antioxidant and antibacterial activities of edible films containing rosmarinic acid were previously demonstrated (Ge et al., 2018). Similarly, Xiong, Kamboj, Ajlouni, & Fang, 2021 and Castro, Andrade, Sanches Silva, Vaz, & Vilarinho, 2019, reported that lipid oxidation of stored fish fillets coated with gelatin-chitosan films containing plant extracts was significantly improved when compared to uncoated samples.

The ability of the developed active films to prevent lipid oxidation in fish samples was also evaluated by the peroxide value (PV) protocol that indicates the amount of the oxidized substances in the samples. As observed in Fig. 4b, and in accordance with the TBARS results, uncoated samples showed a sharp increase in the PV value after the storage time, which can be ascribed to the formation of hydroperoxides. In contrast, the lipid oxidation of the coated salmon fillets was significantly lower, samples coated with PoE-P films being those with the lowest PV value after 8 days of storage. The higher efficiency is attributed to the higher antioxidant capacity of the extracts (Orqueda et al., 2020). Furthermore, the phenolic compounds in the pectin samples might act as a barrier for UV–visible light and oxygen (Fig. 2), displaying a positive impact on lipid photo-oxidation of the salmon fillets (Tzima et al., 2021). Although a slight decrease in the PV value was observed for the AnE-P and PoE-P treatments at 3 and 6 days (when compared to the time 0), it was not statistically significant ($P < 0.05$).

The content of free thiol groups in samples is a commonly used method to determine the degree of oxidation of proteins on foods (Lund, Heinonen, Baron, & Estévez, 2011). The thiol groups of cysteine residues are vulnerable to free radical attack, leading to a wide range of oxidative changes. The results, summarized in Fig. 4c, show that the content of thiol groups in uncoated salmon fillets gradually decreased during the storage period, reaching these values of 10.68 μM thiol/mg protein after 8 days of storage. Thus, these evidenced that these samples showed high

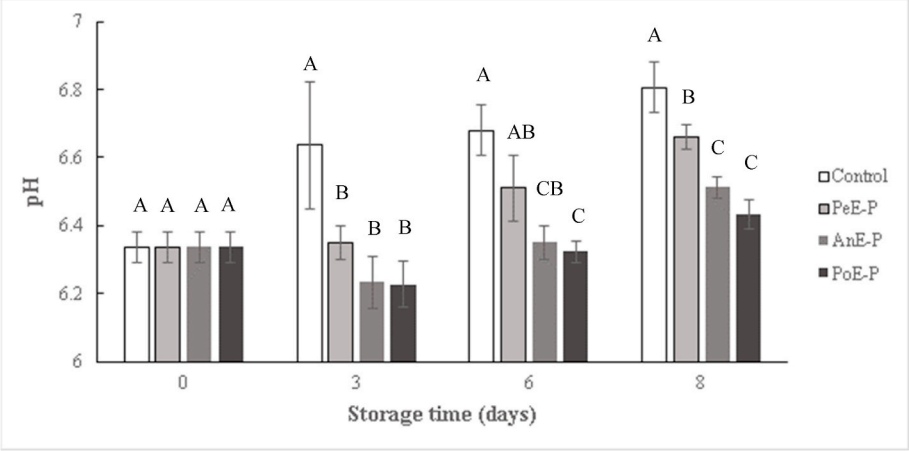

Fig. 5. Changes in pH value of salmon fillet samples during cold storage (4 °C) for 8 days. Films of pectin of red chilto peel (PeE-P) and containing anthocyanins extract from red chilto peel (AnE-P) and polyphenols extract from red chilto peel (PoE-P).The value on day 0 was the initial pH of fish (control), and the bar was repeated on day 0 for clarity and comparison. Bars with differentletters within the same day are significantly different at $P < 0.05$.

protein oxidation. Among the coatings, all treatments with pectin films were effective to prevent protein degradation keeping the content of thiol groups above 15 μM thiol/mg protein during the storage period. After 8 days of storage, the PoE-P sample presented a significantly ($p < 0.05$) higher value of thiol groups compared to the other coated samples (Fig. 4c). This result agreed with the greater antioxidant activity of the PoE-P extract which can be ascribed to the presence of phenolic compounds derived from rosmarinic and caffeoylquinic acids.

3.3.1. *Effect of bioactive films on fish pH*

Changes in pH can be an indicator of spoilage in fish products. The pH values of salmon fillets packed with and without the pectin-based films stored at 4 °C are shown in Fig. 5. The initial pH value for salmon was 6.33, which is within the normal pH range for raw salmon fillets (He, Wu, & Sun, 2014). Throughout the storage period, the pH values tended to increase significantly in the uncoated control sample ($P < 0.05$) (Fig. 5) which results from the enzymatic degradation of proteins into undesirable compounds (i.e., ammonia, trimethylamine, or histamine, among others) (Li et al., 2017). All studied films were able to prevent the increase in pH values over storage. The resulting pH of the samples covered by chilto films was significantly ($P < 0.05$) lower than those of the control sample from day 3 of storage, and this difference even increased on the last day of storage. A higher effect was observed with the AnE-P and PoE-P films, probably due to the content of bioactive compounds in their composition, showing no significant ($P < 0.05$) differences in their effects on the last day of storage. At lower pH, a better microbial inhibition and consequently longer shelf-life of the fish samples is expected (Fan et al., 2009). Similar results in pH were reported for salmon fillets covered by chitosan and gelatin films (Merlo et al., 2019; Xiong et al., 2021). Furthermore, active edible films formulated with seaweeds and designed to preserve fish burgers succeeded in controlling pH changes and microbial growth (Albertos, Martin-Diana, Burón, & Rico, 2019).

4. Conclusions

Chilto, a native species tree of the South American Andean region, is an underutilized species, even though it has demonstrated therapeutic and nutritional values. In this work, pectin-based antioxidant films were developed with the incorporation of polyphenols and anthocyanins enriched extracts into pectin, all of which were obtained from the peel of *S. betaceum* fruits. The active properties of the films were proved on salmon fillets, showing a high antioxidant capacity and good mechanical and physicochemical properties. Among films developed, PoE-P films exhibited a higher ability to protect salmon fillets from oxidation and improve barrier properties. Furthermore, the addition of the extracts to the pectin dispersion did not significantly modify the internal morphology of the films or the thermal properties. Films based on polysaccharides and polyphenols of red chilto peel are interesting as edible coatings to replace non-biodegradable plastics and extend the shelf-life of fish foods. In view of a potential commercialization, it will be necessary to evaluate a sensory analysis through a panel of tasters in order to analyse possible changes in the organoleptic characteristics of the salmon samples for their acceptability.

CRediT authorship contribution statement

María Eugenia Orqueda: Methodology, Investigation, Writing – original draft. **Daniel A. Méndez:** Writing – original draft. **Antonio Martínez-Abad:** Supervision, Formal analysis, Writing – review & editing. **Catiana Zampini:** Writing – review & editing. **Sebastian Torres:** Review. **María Inés Isla:** Funding acquisition, Writing – review & editing. **Amparo López-Rubio:** Conceptualization, Methodology, Funding acquisition, Project administration, Writing – review & editing. **María José Fabra:** Conceptualization, Methodology, Supervision, Formal analysis, Funding acquisition, Project administration, Writing – review & editing.

Declaration of competing interest

The authors have no conflicts of interest.

Acknowledgements

This research was supported by grants from ICOOP-CSIC (COOPB20539), Consejo de Investigación de la Universidad Nacional de Tucumán (PIUNT 2018- G637 Project), Agencia Nacional de Promoción Científica y Técnica (PICT 2017–4416 Project: PICT 2020–3619 Project), and Consejo Nacional de Investigaciones Científicas y Técnicas (PUE 2018–0011 Project), Biolates network "P320RT0186— Sustainable use of Ibero-American vegetable biomass resources in cosmetics" (CYTED) and CSIC Interdisciplinary Thematic Platform (PTI+) Interdisciplinary Platform for Sustainable Plastics towards a Circular Economy+. (PTI-SusPlast+). D.A. Mendez is supported by the Administrative Department of Science, Technology and Innovation (Colciencias) of the Colombian Government (783–2017).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodhyd.2022.107888.

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