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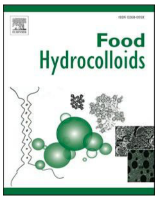

Edible hydrosoluble sachets produced with food by-products and natural additives

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ABSTRACT

In this work, edible films were produced from orange pectin and broken rice (food by-products), combined with different natural additives, namely orange oil, zinc oxide, rice husk (RH) and calcium acetate (CA). The films obtained were fully characterized, presenting similar colour, opacity, thickness and thermal properties. Water contact angles (WCA) ranged from 55 to 85°; water vapor sorption (WVS) between 17.0 and 32.5% (w/w); with a maximum water vapor permeability (WVP) of 14.52×10^{-11} g $m^{-1}.s^{-1}.Pa^{-1}$; Young's modulus between 0.88 and 3.0 GPa; tensile strength (TS) ranging from 6.0 to 19.0 MPa; elongation at break between 0.88 and 3.0%, and almost complete dissolution in water at 60 °C after 15 min. The WCA and mechanical properties of the films were compared with non-commercial and commercial biodegradable films. Orange pectin was the raw material that most influenced the films' properties. PCA (pectin and calcium acetate) and $POO_{1.0}RH$ (pectin, orange oil and rice husk) films showed the best mechanical and barrier properties, being applied as hydrosoluble sachets for white sugar. The organoleptic and physicochemical properties of the sachets were evaluated through sensory analysis and a 12-week shelf-life experiment, respectively. A high level of acceptability was obtained for the sugar sachets, with their physical structure and appearance practically unchanged throughout the tested shelf-life. Edible films obtained from these food by-products and natural additives were shown to be promising alternatives to synthetic sachets for use as sustainable food packaging.

1. Introduction

Growing ecological concerns regarding plastic pollution have led to extensive research into more sustainable alternatives in food packaging. In this case, paper is commonly used to replace plastic in food packaging, since it comes from renewable resources, is biodegradable and recyclable. However, plastic bags generate 39% less greenhouse gas emissions than uncomposted paper bags and 68% less than composted paper bags, require 6% less water, 71% less energy to be produced and generate five times less solid waste (Chaffee & Yaros, 2007). Understanding the disadvantages of plastic and paper as food packaging materials, interest in edible packages has risen in recent years due to their harmless nature to both humans and the environment (Ncube et al., 2020).

Edible materials can be produced from natural and sustainable compounds using modern material-forming technology, have excellent biocompatibility and biodegradability, fulfil the basic functions of food packaging and avoid pollution from plastic packaging. Edible packaging can be made from a variety of food waste and by-products, reducing food waste on an industrial scale and working towards a more circular and sustainable economy. Furthermore, these raw materials are safe for human contact and may contain antioxidant, antimicrobial and dietary compounds that can improve the sensory quality and nutritional value of food products (Zhao et al., 2021).

The three most common categories of edible packaging raw materials are polysaccharides, proteins and lipids, with the first being the most

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widely used due to its abundance in nature, low processing costs and ability to protect food. Compared with traditional packaging materials such as paper or plastic, edibility and ecological performance are two main advantages of polysaccharide-based materials (Zhao et al., 2021). Starch and pectin are among the most widely used biopolymers for the production of food polymer films. Starch-based films are homogeneous, tasteless, semi-permeable to gases, moisture, lipids and aromas (Kong et al., 2022), while pectin-based films present excellent mechanical properties and high water vapor permeability (WVP), which prevents dehydration of the polymer films produced (Chakravartula et al., 2019). However, the films obtained still have poorer mechanical and barrier properties than conventional plastic packaging. Thus, plasticizers, cross-linking agents, fillers, emulsifiers or lipids can be used to improve the films' physicochemical properties (Chakravartula et al., 2019; Kong et al., 2022; Zhao et al., 2021).

In addition to glycerol used as plasticizer (Otoni et al., 2017), essential oils can also be used for their plasticizing effect, enhancing barrier properties of the films, antimicrobial and antioxidant activities (Breceda-Hernández et al., 2020). Other additives have also been used to improve the mechanical and barrier properties of the produced films (Costa et al., 2018; Laftah & Wan Abdul Rahman, 2021; Praseptiangga et al., 2021), increasing the films' flexibility (Colussi et al., 2017), as well as their antimicrobial activity and UV light absorption capacity.

Considering the properties and advantages of using edible films as food packaging, these materials are preferred in the production of various types of packaging such as soluble pouches, bags, packets or sachets to contain powdered substances such as seasoning and flavouring additives for foods, instant coffee, milk powder, tea leaves, and other food additives (Liu et al., 2020).

In this work, orange pectin and broken rice were used as food by-products in the production of edible films, incorporating additives of mineral (calcium acetate (CA)), vegetal (rice husk (RH) and orange essential oil (OO)) and metallic (zinc oxide (ZO)) nature in the formulations to improve their physicochemical properties. The raw materials used, and films produced were fully characterized and the better performing films were used as a proof of concept for the development of hydrosoluble white sugar sachets to replace the common paper bags used in this type of good. The organoleptic properties of the sachets were assessed by sensory analysis and the physicochemical properties by shelf-life testing.

2. Materials and methods

2.1. Chemicals and raw materials

All used equipment, agri-food wastes and by-products were sanitized, being these raw materials processed as reported in our previous work (Jorge et al., 2023). Orange pectin (P) and orange oil (OO) were extracted from orange peels, provided by local food producers in the region of Coimbra (Portugal), using the procedure detailed by Hosseini et al. (2016) and hydrodistillation, respectively. Eggshells, a source of calcium carbonate ($CaCO_3$) for the production of calcium acetate (CA), were provided by "Derovo - Derivados de Ovos, S.A." (Pombal, Portugal). Broken rice and RH were provided by food industries from the "Agricultural Cooperative of the Municipality of Montemor-*o*-Velho" (Coimbra, Portugal).

Glacial acetic acid (AA) was purchased from Honeywell (Fluka, $\geq$99.8%), zinc oxide (ZO) from Fagron (Netherlands, $\geq$99.8%), commercial starch, commercial pectin from citrus peel and glycerol from Sigma-Aldrich (USA). Sterilized water and Milli-Q water were used to prepare the filmogenic mixtures and measure the water contact angles (WCA), respectively.

The CA additive was produced by dissolving eggshells (composed of 95% $CaCO_3$ (Ajayan et al., 2020) in a 10% (v/v) aqueous solution of glacial acetic acid (AA)). This concentration of AA has been reported as optimal in the production of biodegradable films by several authors (Andrade Martins et al., 2020; Wu et al., 2021). The CA solution obtained was then added to the filmogenic mixture until the amount of calcium ions (Ca^{2+}) present in the mixture was equal to the amount of Ca^{2+} present in a 1 % (w/w) solution of calcium chloride, a salt most commonly used and reported in the manufacture of biodegradable films (Costa et al., 2018). The incorporation of CA in the orange pectin-based films of this work aims to provide a crosslinking enhancement induced by the Ca^{2+} and the pectic chains, forming the so called egg-box model usually obtained with alginate-based films (Costa et al., 2018). Since both pectin and alginate are polysaccharides obtained by repeating units of sugar molecules, these two compounds can bind effectively to Ca^{2+} molecules and lead to improved crosslinking between the polymeric chains of the films' phase-forming compounds. This results in the formation of films with higher mechanical strength and stability. The pectin's degree of esterification (DE) is crucial for determining the calcium-driven interactions that will enhance the films' physicochemical properties, as pectins with a DE inferior to 50% tend to have fewer methoxyl groups available for calcium cross-linking, relying more on non-calcium-ion-driven interactions to provide structural integrity to the films' polymeric structure, such as hydrogen bonding and hydrophobic interactions, and leading to films with lower mechanical properties (comparing to films obtained by using highly esterified pectins). Conversely, highly esterified pectins (DE above 50%) possess more methoxyl groups available for calcium binding, facilitating stronger interactions with calcium ions and the formation of a networked structure, which leads to the production of films with improved mechanical and barrier properties. However, it should be noted that pectins with slight variations within this range (*e.g.*, pectin with 45% DE versus pectin with 55% DE) may exhibit comparable behaviour due to the inherent variability in food matrix composition, with other factors being also pivotal in dictating the specific interactions and functionalities of pectin-containing films. The calcium ion concentration, temperature, pH and the presence of other additives must be carefully analyzed as well, as too high or too low values of each of these parameters can reduce the mechanical and barrier properties of the films.

In this work, as the CA solution was produced considering the optimal concentrations of Ca^{2+} and AA reported in previous works, it is expected that the obtained films' properties present good mechanical and barrier properties when CA is incorporated in the formulation.

The contents of raw fiber, protein, starch, lipids and ash in broken rice and rice husks, shown in Table 1, were determined as described in the previous work (Jorge et al., 2023).

The purity of the extracted pectin was evaluated based on the titration of deesterified pectins after acid precipitation, with six drops of phenol red (0.02% (w/v)) as an indicator, instead of the Hinton's indicator mentioned in the adapted procedure (Sheukhina & Fedichkina, 1994). The DE of pectin was calculated using a Fourier Transform Infrared (FTIR) method (Kyomugasho et al., 2015). Both results are expressed in Table 1.

Table 1
Broken rice, rice husk, extracted orange pectin and commercial pectin's characterization.

By-products/Raw materials	Raw fibers (%)	Protein (%)	Starch (%)	Total sugars (%)	Lipids (%)	Ash (%)
Broken rice	0.37 ± 0.08	10.90 ± 0.35	60.50 ± 2.76	87.63 ± 0.35	0.50 ± 0.11	0.70 ± 0.02
Rice husk (RH)	48.52 ± 0.30	2.47 ± 0.05	19.40 ± 0.16	1.22 ± 0.03	1.22 ± 0.02	13.55 ± 0.48
Pectin		**Purity (%)**		**Degree of esterification (%)**		
Extracted from orange peels		77.72 ± 0.58		54.76 ± 2.76		
Commercial		90.91 ± 5.96		53.64 ± 1.43		

2.2. Edible films production

Considering the results from our previous work (Jorge et al., 2023), the formulation selected as the reference in this study was the one coded as P$_P$O$_{PC}$, whose base formulation was orange pectin and broken rice 1.2% (w/v) each, glycerol and commercial starch 0.4% (w/v) each. In the present study, this formulation was called P and therefore did not include any of the additives tested (Table 2). To improve the functional properties of the films, additives of different origins were introduced into the base formulation (see section 2.3), as detailed in Table 2, up to a total amount of starch of 1.13 g (obtained by accounting the composition of the broken rice defined in the previous work (Jorge et al., 2023) and the amount of commercial starch added).

The film codes were then differentiated according to the additives used in each film and the amount of orange oil (OO) they contained. For example, POO$_{0.5}$RH is a film containing all the raw materials of the reference formulation in their respective proportions, together with 0.5% (v/v) of OO and 1% (w/v) of RH, whereas POO$_{0.5}$CA is the same composition with 1.0% (v/v) of CA added instead of RH. All the films were prepared and stored, as reported in the previous study (Jorge et al., 2023).

2.3. Additives' incorporation

The influence of 0.5 and 1.0% (v/v) of OO on the properties of the films was evaluated, with 1.0% (v/v) being the maximum amount of OO that can be used without compromising the organoleptic properties of the films (Coimbra et al., 2023). The impact of adding 1 g of ZO, RH and CA per 100 g of the total amount of P and starch present in the filmogenic mixture was also studied. The amount of ZO used in the formulations was based on several research papers (Praseptiangga et al., 2021; Roy & Rhim, 2020) which reported 1% (w/w) as the optimum concentration, considering the total film-forming polymers present in the mixture. This percentage was also used for RH and CA to compare the influence of all additives on the properties of the films obtained.

2.4. Physicochemical characterization of the edible films

The chemical composition of each film was assessed by applying attenuated total reflection – Fourier transform infrared spectroscopy, ATR-FTIR (PerkinElmer, Universal ATR sampling accessory, UK) to the film surfaces as well as to the individual raw materials. The experiments were performed at 64 scans with a 4 cm^{-1} resolution, between 550 and 4000 cm^{-1}.

The thermogravimetric properties were evaluated by differential scanning calorimetry/thermogravimetric analysis, DSC/TGA (TA Instruments, model Q600, USA). Replicated samples were heated at 10 °C/min, from 25 °C to 600 °C, under a nitrogen atmosphere (100 mL/min) as detailed in the previous work (Jorge et al., 2023).

Table 2

Amount of the additives included in the base formulation (orange pectin/broken rice/commercial starch/glycerol) per 100 mL of suspension.

Film code	Additives$^{\bullet}$				Solid content (g)
	Orange oil (OO) (mL)*	Zinc oxide (ZO) (g)*	Rice husk (RH) (g)*	Calcium acetate (CA) (mL) *	
P	0	–	–	–	2.80
PZO		0.023	–	–	2.82
PRH		–	0.023	–	
PCA		–	–	2.44	2.80
POO$_{0.5}$	0.5	–	–	–	2.80
POO$_{0.5}$ZO		0.023	–	–	2.82
POO$_{0.5}$RH		–	0.023	–	
POO$_{0.5}$CA		–	–	2.44	2.80
POO$_{1.0}$	1.0	–	–	–	2.80
POO$_{1.0}$ZO		0.023	–	–	2.82
POO$_{1.0}$RH		–	0.023	–	
POO$_{1.0}$CA		–	–	2.44	2.80

*values calculated as described in section 2.1.

To assess the films' colour parameters in the CIEL*a*b* colour system, a Minolta CR-200b colourimeter (Japan) was used, performing at least five random measurements for each film. The films were evaluated in terms of lightness L* (where 0 = black to 100 = white), a* (where −60 = green to +60 = red) and b* (where −60 = blue to +60 = yellow). The chroma (C*) and hue angle (h*) of the films were also determined. Film opacity was measured using a UV-VIS spectrophotometer (UV-VIS Jasco V-530 Spectrophotometer, Japan). The film samples were cut into strips (4 cm × 1 cm) and placed in glass cuvettes, with an empty cuvette as a reference. The results were calculated using **Equation (1).**

$$\text{Opacity}\left(\text{mm}^{-1}\right)=\frac{A_{600}}{l} \tag{1}$$

Where A$_{600}$ is the absorbance of the sample at 600 nm and l is the thickness of the films in mm. Measures were made in triplicate.

The thickness of the films was measured using a digital micrometer (Tesa Micromaster Electronic Micrometers IP54) with an accuracy of 0.001 mm, with six measurements being taken at random locations on each film.

Films were characterized according to their WCA and WVP using the procedures detailed in the previous work (Jorge et al., 2023). To evaluate water vapor sorption (WVS), four samples with 1 cm^2 of each film were cut and stored in a desiccator with magnesium nitrate ($Mg(NO_3)_2$), maintaining 55% of relative humidity at room temperature (25 °C). The mass of each sample was recorded after 12 and 24 h, to ensure that all samples reached equilibrium, presenting the maximum WVS possible in that environment. Once the recorded mass was constant, films were placed in a controlled atmosphere chamber (Binder KBWF 24, Germany) and maintained at 80% relative humidity at 25 °C for 10 days, weighed twice a day. When equilibrium was reached again, the samples were dried in an oven at 80 °C (Venticell, Germany) for 3 days and measured twice a day, until their mass became constant. **Equation (2)** was used to determine the WVS of the films at 80% relative humidity.

$$\text{WVS}\ (\%, \text{w}/\text{w})=\frac{m_{Relative\ Humidity}-m_0}{m_0} \tag{2}$$

Where m$_{\text{Relative Humidity}}$ is the final mass of the sample at 80% relative humidity, and m$_0$ is the mass of the dried sample.

Young's modulus (E′), tensile strength (TS) and elongation at break (ETB) were measured using the Inspekt mini (Hegewald & Peschke) equipment, with a 3 kN load cell at a speed of 5 mm/min, according to the same standard and procedure used in our previous work (Jorge et al., 2023) to evaluate mechanical properties of the films.

The water dissolution (WD) of the films was also recorded, and the shelf-life experiment was carried out to assess the potential of these films in the marketplace. A new procedure was used to determine the WD of the films. Three samples with 2 mg of each film were dissolved in quartz cuvettes with 2 mL of bi-distilled water (Milli-Q water) at two different temperatures (30 and 60 °C), under magnetic stirring at 200 rpm, for 60 min. For this test, a UV-VIS Jasco V-650 spectrophotometer (Japan) was used, being able to maintain the correct temperature and agitation during all experiments. The absorbance of the samples at 550 nm was recorded every 3 min. This wavelength was selected since it is commonly used in monitoring and controlling the quality of drinking water (Shi et al., 2022). The turbidity of each dissolved film was calculated using Equation 3(Narayanan et al., 2006).

$$Turbidity\left(g^{-1}\right)=\frac{2.3\bullet A_{550}}{m\bullet L} \tag{6}$$

Where A$_{550}$ is the absorbance of the sample at 550 nm, m is the mass of the sample dissolved in the cuvette in g, and L is the optical length of the cuvette (1 cm) (Narayanan et al., 2006).

The collected data to characterize the films and raw materials are expressed as mean values ± standard deviation, being subjected to analysis of variance (one-way ANOVA) and the means being compared through the Tukey test (95% confidence level) in JMP Pro 17 statistical software.

2.4.1. Sugar sachets preparation

The two edible films with the most suitable properties to be applied as sugar sachets, considering the physicochemical properties of the films evaluated in this study, were used to produce water-soluble sachets. Two pieces of edible film (2 × 2 cm) were cut and glued together at the edges using carboxymethylcellulose-based glue (0.03 g/mL) with sterilized water. Approximately 350 mg of white sugar was inserted into each sachet, corresponding to an 11.5-fold reduction compared to

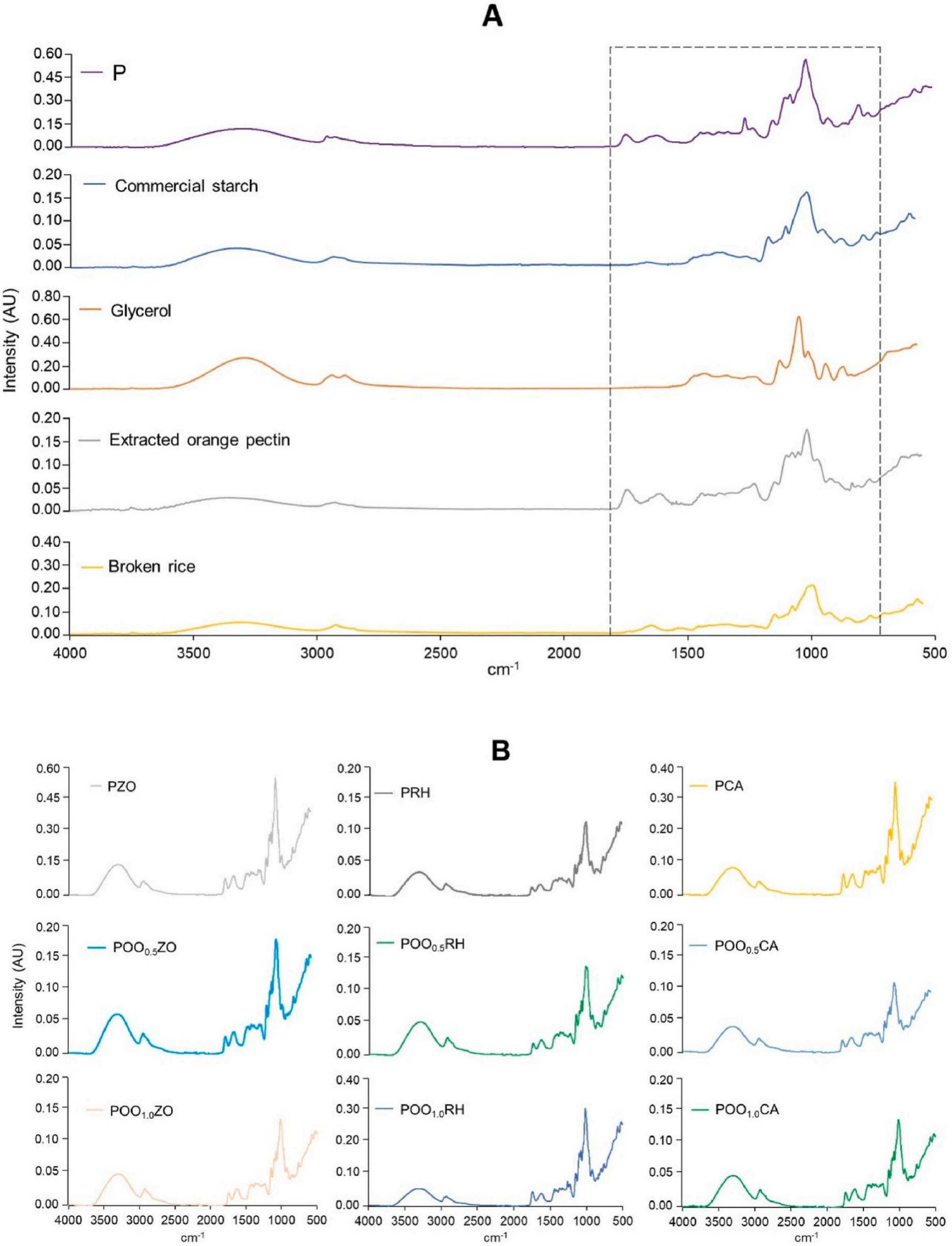

Fig. 1. FTIR spectra of P and the main raw materials used to produce it (**A**), and FTIR spectra of all the films with additives (**B**).

commercial sugar sachets (original dose of 4 g/sachet).

2.4.2. Sensory analysis

For sensory evaluation of the sachets, a total of 35 untrained volunteer panellists were invited to give their opinions on two different question-type situations. The first on the acceptability of tea samples prepared with edible sugar sachets compared to the usual lemon balm tea. The second is to give their opinion on the physical appearance of the sugar sachets, sugar-consuming habits and possible use of this type of edible hydrosoluble sachets in everyday life.

A procedure of data analysis was defined with the sensorial form prepared to be answered anonymously, to protect the rights and the privacy of volunteers, who consented to provide information for their minimum identification (such as gender and age) and to participate in this research.

The acceptability test consisted of questioning the panellists about the use of the hydrosoluble sugar sachets in 30 mL of lemon balm tea with a final dissolved sugar concentration of 10 g/L, with or without hydrosoluble sachets. The amount of white sugar per 250 mL of tea (teacup) was defined as 2.5 g, which corresponds to the addition of ~63% of a 4 g sugar paper sachet in a cup of tea. The mass of edible film added to the tea was necessary to maintain the same proportion between white sugar and edible film used in the production of hydrosoluble sugar sachets (*i.e.*, for every 350 mg of white sugar added, approximately 50 mg of PCA and 65 mg of $POO_{1.0}RH$ were added to the mixture). Panellists are asked about the aroma, colour, homogeneity and flavour of the tea samples, their acceptability, whether the amount of added sugar was in line with their personal preferences or whether they would prefer a higher or lower amount of sugar in tea.

In the second questionnaire, panellists had to indicate whether they use sugar daily, whether they would buy sugar hydrosoluble sachets, whether the colour and appearance of the sachets were acceptable and whether transparency is a necessary property for this type of product.

The sensory analysis was carried out at the Department of Chemical Engineering at the University of Coimbra (Portugal), with essential safety procedures to guarantee the integrity of the results and the well-being of the participants.

2.4.3. Ethical approval

Ethics approval was not required for this research.

3. Results and discussion

3.1. Chemical composition of the raw material and film surface

The evaluation of the chemical modifications occurring between the components of the films was assessed through spectroscopic analysis applied to the samples (raw materials and films).

All spectra are in Fig. 1A showed a broad peak around 3350 cm^{-1} due to OH stretching, and two smaller peaks around 2930 cm^{-1} and 2880 cm^{-1} from C–H vibrations of methyl esters (Chakravartula et al., 2019). It is observed that orange pectin is the raw material with the greatest influence on the spectrum of the P film, as the peaks of the film spectrum are very similar to the spectrum obtained for this by-product.

Even so, it is observed that the main peak of P film, in the range of 1200 and 800 cm^{-1}, has two small shoulders on the left side, a very sharp peak in the middle, a smooth decrease on the right side and a small shoulder at the end. The profile of the two small shoulders on the left is very similar to those found in the spectra of commercial starch and broken rice, with the sharp peak being very similar to that found in the spectrum of glycerol. The smooth decrease on the right with the small shoulder at the end, also more closely resembles commercial starch and broken rice, as glycerol and pectin have a small shoulder in this region of their spectra. Therefore, although pectin is a very important material for the formation of the film structure, the other components seem to play a very important role as well.

Comparing the spectrum of film P with that of the other films produced, no significant changes were observed for the films with and without additives, as can be seen in Fig. 1B. However, it was observed that in some spectra of films with additives, the peaks around 1740 cm^{-1} (C=O bonds of esters) (Chakravartula et al., 2019) were less prominent. This trend is easily observed in films where OO has been incorporated, although it is also found in films with other additives. As this peak is one of the peaks associated with orange pectin and is used to obtain the degree of esterification of this material, its less prominent and broader profile may be related to possible bonds of the additives with COOH and $COOCH_3$ groups of pectin (Coates, 2006; Vaezi et al., 2019). The main influence of pectin in the films' structure and physicochemical properties is related to its high degree of esterification (greater than 50%, being approximately 55% as shown in Table 1). The addition of high methoxyl pectin (HMP) predominantly induces significant crosslinking among pectic chains and other film-forming compounds. Due to the abundant –OH groups in their structure, starch-rich materials like commercial starch or broken rice, can interact with the COOH and $COOCH_3$ groups of pectin and modify the films' physicochemical properties.

The presence of carbohydrates with low molecular weight distribution in broken rice (about 1/3 of the carbohydrates of this ingredient are of a low molecular weight, Table 1) can also reduce the ability of the tested additives to effectively alter the films' structure. These small-size compounds might have the ability to fill up the open spaces within the films' polymeric structure and lead to a competition with the additives investigated for those sites, namely plasticizing compounds such as OO. Thus, whether by reducing the interactions between the additives and pectin by the abundant –OH groups in its structure or by limiting the number of open spaces within the films' structure where plasticizers can be introduced, broken rice can mitigate the influence of additives in the films' structure and have a major impact on the films' physicochemical properties. This might explain the similarities observed between the produced films, as the small amount of additives added to the film formulations and the facilitated interactions between the film-forming compounds can reduce the influence of those additives in the films' properties and limit the impact of some of the system's variations investigated.

3.2. Thermal properties of the films

The films' thermograms are presented in Fig. 2 and are compared with the raw material thermal events reported in previous work (Jorge et al., 2023).

All films presented similar thermograms, with the same number of thermal events and identical temperature peaks. The initial peak observed is associated with the loss of free and bound water. The main degradation peaks were observed at 225 °C and/or 300 °C since several films showed similar intensity in both peaks. As the first peak of degradation appears around 225 °C for all films, it can be stated that even though the main peak of each one appears at different temperatures, they all have similar thermal properties (Chakravartula et al., 2019).

When comparing the thermograms of the main raw materials with the films obtained, it was possible to observe their similarity with those of P film, with the peak temperatures being very close to each other, as observed in the example of Fig. S1 (Supplementary Data). The significance of the orange pectin in the final polymeric structure of the films is shown, and the authors reported the same trend for the use of HMP in edible films (Shafie et al., 2020).

Thus, considering the results obtained from the surface chemical composition and thermogravimetric compositions of the films and their originating raw materials, it is possible to state that the extracted orange pectin incorporated in all filmogenic mixtures had a great influence in the films' polymeric structure, with a minor (but important) interference from other film-forming compounds and additives as also confirmed.

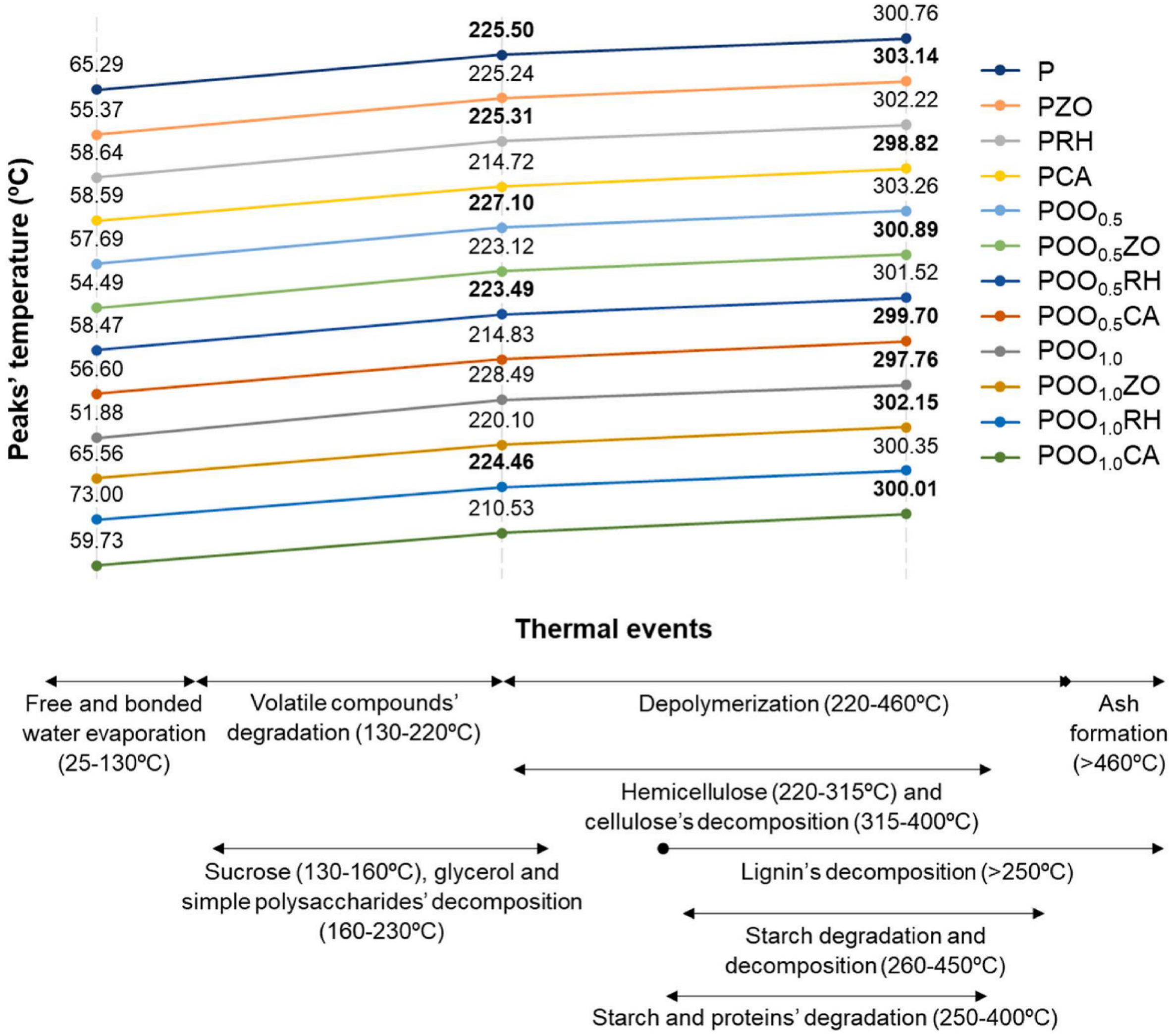

Fig. 2. Peaks' temperatures and possible thermal events associated with them.

3.3. *Colour, opacity and thickness of films*

Film colour is an important physical property as it directly affects the appearance of products. Typically, progressively brighter films are classified as higher-quality films (Ulfah et al., 2018).

The films' colour parameters, L*, a* and b*, are presented in Table 3. For all films, the luminosity (L*) was very high, ranging from 94.49 ± 0.23 to 96.72 ± 0.03. Film P was the film with the highest luminosity value, indicating that the presence of additives in the formulation reduced L* in the films produced. The use of ZO led to the lowest L* values, with other authors reporting the same trend and relating it to the light barrier properties of this compound (Motelica et al., 2021; Vaezi et al., 2019). Among films with additives, in general, those with CA showed a higher L*, followed by films with OO, RH and ZO. This agrees with previous works, where it was reported that films produced with RH and CA have good transparency/luminosity (Elango et al., 2016; Kumar et al., 2010; Rabbani et al., 2022). The incorporation of OO into films with any additives reduced the L* value, this phenomenon is associated with the yellowish colour of OO (Escamilla-García et al., 2017). The L* values for all films in this study are in line with the highest values present in literature, giving the films good optical properties (Dou et al., 2018; Escamilla-García et al., 2017; Ulfah et al., 2018).

Concerning the colour parameters a* and b*, it was observed that the use of additives in the films reduced a* and increased b*. However, their range of values lies in a narrow achromatic region of the CIELAB's colour space, with a* ranging from −1.0 to 0.0 and b* from 2.0 to 10. CA is the additive that most reduce a*, followed by ZO, with RH and OO being those that least influence a* when compared to P film. The variations in a* values are relatively small and do not significantly change the colour of the films. Films with ZO had a higher b*, making them more yellow and in line with previous reports (Motelica et al., 2021). The variations in b* are not very significant in the optical properties of the films. Therefore, the films showed very good luminosity and similar colourimetric parameters.

The opacity of the films (Table 3) was also studied to understand the effect of each additive on their transparency, as a film with higher opacity has lower transparency. Transparency is a very important feature in the application of edible films, as it allows the protected goods to be seen (Chakravartula et al., 2019). Although the opacity values varied between 2.0 and 7.0 mm^{-1}, all films showed high transparency. These values follow previous work where edible films as food packaging have been shown to have opacities between 0.28 and 8.41 mm^{-1} (Chakravartula et al., 2019; Shahrampour et al., 2020; Vaezi et al., 2019).

The film with the highest opacity was P, therefore the least transparent compared to all films with additives that showed higher transparency. This is probably due to the introduction of new compounds into the polymeric matrix, which can improve film component compatibility (Zuo et al., 2019). As the pectin used in this study is HMP, the higher opacity in the films without additives may be associated with higher

Table 3
Colour parameters (L*, a* and b*), opacity and thickness of the produced films.

Film code	L*	a*	b*	Opacity (mm^{-1})	Thickness (μm)
P	96.72 ± 0.03^{a,b}	−0.15 ± 0.03^{a,b}	2.14 ± 0.09^{q}	6.80 ± 0.02^{a}	43.67 ± 2.75^{i,j,k,l}
PZO	94.49 ± 0.23^{m}	−0.55 ± 0.05^{h,i,j}	9.12 ± 0.39^{a}	2.80 ± 0.00^{j}	48.91 ± 3.78^{b,c,d,e,f,g}
PRH	95.92 ± 0.10^{e,f,g,h,i}	−0.17 ± 0.02^{a,b,c}	3.41 ± 0.10^{j,k,l,m,n}	3.87 ± 0.01^{i}	48.85 ± 2.82^{b,c,d,e,f,g,h,i}
PCA	96.08 ± 0.22^{d,e,f,g}	−0.59 ± 0.03^{j,k}	5.00 ± 0.32^{e,f,g,h}	2.84 ± 0.02^{j}	49.84 ± 4.80^{b,c,d,e}
POO$_{0.5}$	95.85 ± 0.03^{f,g,h,i}	−0.19 ± 0.02^{a,b,c,d}	3.30 ± 0.06^{j,k,l,m,n,o,p}	4.47 ± 0.01^{f}	45.86 ± 2.26^{d,e,f,g,h,i,j,k,l}
POO$_{0.5}$ZO	94.79 ± 0.09^{l,m}	−0.39 ± 0.01^{f,g}	6.93 ± 0.20^{b}	4.33 ± 0.05^{g}	49.56 ± 3.04^{b,c,d,e}
POO$_{0.5}$RH	95.22 ± 0.13^{j,k,l}	−0.21 ± 0.00^{a,b,c,d}	4.68 ± 0.24^{f,g,h,i}	5.45 ± 0.05^{c}	50.28 ± 3.56^{b,c,d}
POO$_{0.5}$CA	96.12 ± 0.26^{d,e,f,g}	−0.64 ± 0.02^{k,l}	4.74 ± 0.57^{e,f,g,h,i}	2.14 ± 0.02^{k}	51.06 ± 2.61^{b,c}
POO$_{1.0}$	95.90 ± 0.17^{e,f,g,h,i}	−0.14 ± 0.01^{a}	3.82 ± 0.15^{i,j,k,l}	4.25 ± 0.00^{h}	48.23 ± 3.04^{b,c,d,e,f,g,h,i,j}
POO$_{1.0}$ZO	94.64 ± 0.08^{m}	−0.36 ± 0.02^{e,f}	7.01 ± 0.23^{b}	5.56 ± 0.00^{b}	52.67 ± 2.72^{b}
POO$_{1.0}$RH	95.45 ± 0.11^{i,j,k}	−0.15 ± 0.00^{a,b}	4.25 ± 0.14^{g,h,I,j}	4.64 ± 0.01^{e}	48.07 ± 2.12^{b,c,d,e,f,h,h,i,j}
POO$_{1.0}$CA	94.95 ± 0.13^{l,m}	−0.44 ± 0.00^{g}	6.80 ± 0.22^{b,c}	4.80 ± 0.01^{d}	58.38 ± 1.66^{a}

*Different letters in the same columns differ significantly ($P < 0.05$) by the Tukey test.

entanglement between the film-forming components, resulting in more crystalline structures (Shafie et al., 2020). The introduction of additives resulted in less ordered structures in the films, reducing their crystallinity and increasing their transparency. Other authors reported the same tendency, with composite films being less opaque than films produced exclusively with the respective film-forming compounds (Shahrampour et al., 2020). Among the films with additives, those with OO presented higher opacity values, with this reduction of transparency associated with the increase in light scattering of the films (Sharma et al., 2021). To facilitate a comparison of the colour and opacity observed in the films, POO$_{1.0}$ and PZO are shown in Fig. 3A.

The thickness of the films ranged from 43.67 ± 2.75 μm up to 58.38 ± 1.86 μm. The results are presented in Table 3. Although the values are similar, it is possible to observe that the films with additives have a higher thickness than the films without any additives (P). This is explained by the intermolecular interactions between the film-forming compounds and the resulting polymeric structure, as the use of additives increases the solid content in the film matrix, making it denser and thicker (Singh et al., 2019). In addition, some of these compounds can reduce the intermolecular interactions between the film-forming compounds in the polymeric matrix (namely between the pectic chains and the pectin and starch chains), creating more free volume within the film structure and increasing its thickness (Shafie et al., 2020). The film with the highest thickness was POO$_{1.0}$CA. All films with CA had some of the highest thickness values. The increased thickness of the films with CA was related to the induced acetylation process of the polymeric chains, Ih led to the introduction of an acetyl group in the chemical structure of the films and promoted larger spaces between the polymeric chains (Colussi et al., 2017). However, as the difference between the film thicknesses was not very pronounced, it was not a relevant parameter for selecting films with better functional properties.

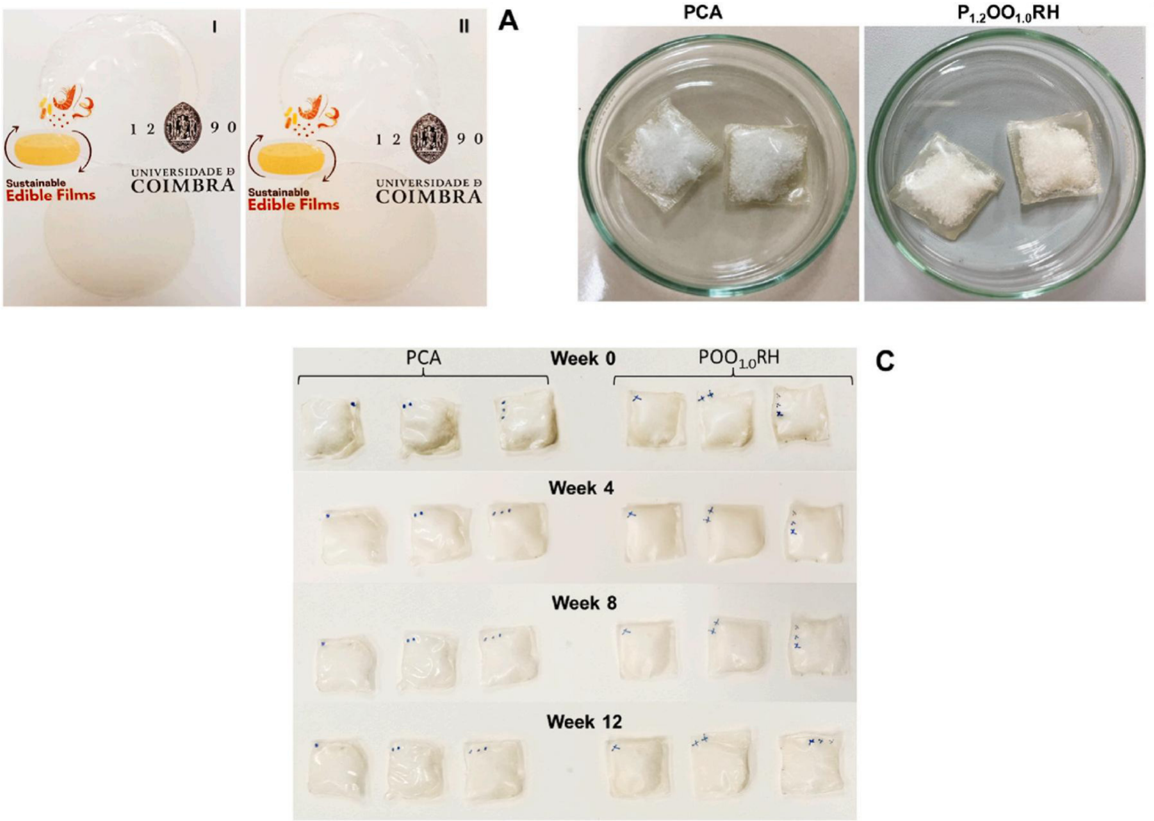

Fig. 3. Comparison between the films with greater colour (**I-A**) and opacity (**II-A**) differences: (**I-A**) POO1.0 (top) and PZO (bottom), and (**I–B**) POO0.5CA (top) and P (bottom); and the visual appearance of the sachets (**B**) during the 12 weeks of sensory analysis (**C**).

3.4. Water contact angle, water vapor sorption and water vapor permeability of films

Wettability is a crucial property of edible films since it aids in comprehending the polymer surface's interactions with water. Measuring WCA enables films' surfaces to be classified as hydrophilic (WCA between 0 and 90°) or hydrophobic (WCA between 90 and 180°) (Ngo et al., 2018).

The WCA values, exhibited in Fig. 4, varied between 55.0 ± 2.65 and 85.20 ± 4.45°. Comparing P with the other films containing additives, it is possible to observe that three of the four films with the highest WCA have CA in their composition. These results are related to the acetylation process, enabling the replacement of hydrophilic chemical groups in the film matrix, such as hydroxyl or carboxyl groups, with acetyl groups, increasing the hydrophobicity of the films (Colussi et al., 2017). All films with ZO are included in the group of films with the lowest WCA values, with other authors reporting the same trend and associating these results with an increase in surface roughness, due to the incorporation of this additive (Ngo et al., 2018). Regarding the addition of OO, it was observed that the incorporation of 1.0% ($POO_{1.0}$) had a significant increase in the WCA and also in the hydrophobicity of the films, but using 0.5% ($POO_{0.5}$) did not significantly change the WCA compared to P. The reduction in hydrophilicity in the films due to the incorporation of OO is related to the hydrophobic nature of essential oils and the corresponding increase in the number of hydrophobic components present in the film matrix (Escamilla-García et al., 2017; Motelica et al., 2021).

Given that most of the obtained WCA values are above 65°, and some are relatively close to 90°, it is possible to conclude that the majority of the films obtained demonstrated a hydrophobic nature. Furthermore, the results fall within the range of previous studies on edible films intended for use as both soluble sachets and various forms of food packaging (between 40 and 83°), with PCA and $POO_{1.0}CA$ demonstrating the highest WCA values in this research (83.88 ± 3.41 and 85.20 ± 4.45°, respectively) (Chakravartula et al., 2019; Zhao et al., 2021).

To assess the WVS of the produced films, their sorption capacity was measured at 80% relative humidity. The results (Fig. 4) varied from 17.67 ± 2.33 and 32.44 ± 2.22 % (w/w) and agreed with other reports on food packaging edible films (Chakravartula et al., 2019; Shafie et al., 2020).

In general, films with additives exhibited higher WVS values than films without additives. The increase in the free volume within the film matrix due to the presence of additives reduces the number and strength of entanglements between pectic chains and other film-forming compounds. As a result, the intermolecular area available for water molecules to diffuse and be absorbed into the polymeric structure is increased, resulting in a high WVS of the films (Shafie et al., 2020). The films with the highest WVS values contain both RH and CA. These results could be attributed to an increase in hydroxyl and carboxyl groups from RH, as well as acetyl groups from CA. These groups increase the affinities towards water molecules (Tongdeesoontorn et al., 2011) and enlarge the intermolecular spacing between the polymeric chains of the films' structure. As a result, the films can retain more water and achieve higher WVS (Colussi et al., 2017). Raising the concentration of OO in the formulations led to a reduction in WVS. This is attributable to the hydrophobic properties of OO, which result in the films having greater hydrophobic character (Escamilla-García et al., 2017; Motelica et al., 2021).

The WVP of the films (Fig. 4) ranged from 10.89×10^{-11} to 14.52×10^{-11} $g\ m^{-1}.s^{-1}.Pa^{-1}$, which falls within the range of WVP values reported for edible films used as packaging by other authors (Vargas et al., 2008). The inclusion of all additives in the film decreased their WVP as compared to the film without any additives (P). The films with lower WVP contained either CA, OO or both of these additives. The results for OO were predictable, as this additive's incorporation in the films' polymeric structure augments the number of hydrophobic compounds, leading to a reduction in water molecule permeability through the films' matrix (Escamilla-García et al., 2017; Motelica et al., 2021). The reduction in WVP upon the incorporation of CA may be associated with the acetylation procedure detailed in this study. The expansion of the polymeric chains enhances resistance to the diffusion of water molecules through the film, leading to mower WVP values (Colussi et al., 2017).

3.5. Mechanical properties of films

The mechanical properties of the films are critical to their use as food packaging. Low strength or lack of elasticity can lead to reduced structural integrity of these materials during production, handling and

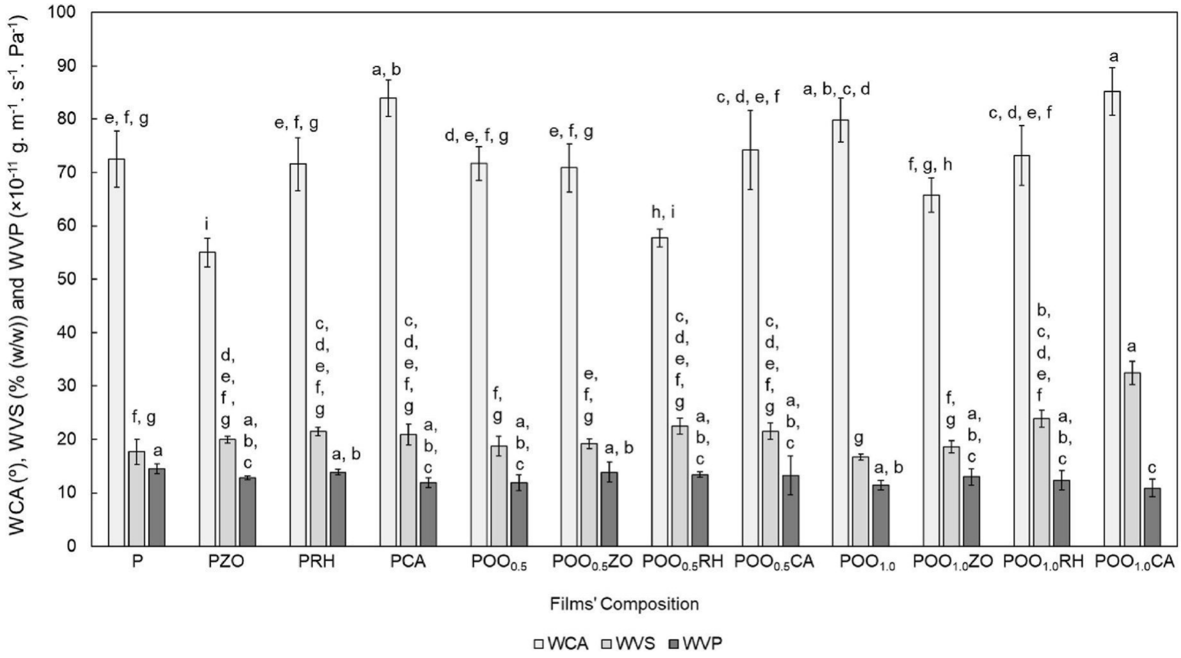

Fig. 4. WCA, WVS and WVP values of the produced films. Columns with different letters differ significantly ($P < 0.05$) by Tukey test.

storage (Shafie et al., 2020). This can cause significant problems and decrease the overall quality of the final product. Therefore, it is essential to carefully consider the properties of the materials and their suitability for the intended use to avoid any potential issues. The results of Young’s modulus (E′), tensile strength (TS) and elongation at break (ETB) for the produced films are shown in Fig. 5.

For this study, the E′ values of the films ranged from 0.88 ± 0.11 to 3.05 ± 0.13 GPa. Compared to P, all films with a single additive exhibited similar or higher values of E’. However, the simultaneous addition of ZO, RH, or CA to films comprising OO led to a decrease in the E′ values. These results suggest that the concentration of non-film-forming compounds is very important for the films’ physicochemical properties. When only one additive is added, the concentration of additives is low, and these molecules cannot significantly reduce the entanglements between the film-forming compounds. Therefore, the additives can only fill the free space within the polymeric structure of the films, promoting hydrogen bonds with the film-forming compounds and causing the films’ stiffening (Lim & Gong, 2018; Ngo et al., 2018). Thus, the expected plasticizing effect of OO is diminished, as these molecules occupy the spaces between the entanglements, restricting the free space and mobility of the polymeric structure, and increasing the rigidity of the films (Lim & Gong, 2018; Shafie et al., 2020).

When the films contain two additives in their composition, the higher amount of non-film-forming compounds reduces the number and strength of the HMP’s entanglements within the polymeric structure, thereby decreasing the rigidity of the films (Shafie et al., 2020). The importance of the additives’ concentration in the film’s rigidity is supported by the high E′ values of the films featuring solely OO (POO$_{0.5}$ and POO$_{1.0}$), and the subsequent decreasing the E′ values when any additional additive is introduced to the films comprising OO.

In terms of ETB, the values ranged between 0.88 ± 0.07 and 2.64 ± 0.32 %. Higher E′ values of a film lead to greater rigidity and lesser flexibility and elongation. Consequently, a film with greater E′ tends to have lower ETB due to the reduced molecular mobility of the polymeric chains (Shafie et al., 2020). This trend is confirmed by the data, as POO$_{1.0}$ displays the highest E′ value and the lowest ETB value, while POO$_{0.5}$ZO has the lowest E’ value but the highest ETB value.

The use of a single additive in the films (*e.g.* OO), as well as increasing its concentration, caused a reduction in the flexibility and corresponding ETB values in the films when compared to those without additives (P). However, the addition of ZO, RH or CA into OO-containing films enhances flexibility and ETB values. This aligns with previous findings for E’, as a higher concentration of additives can decrease intermolecular interactions among film-forming compounds. This increases free volume within the polymeric matrix and molecular mobility of produced films, resulting in higher ETB values (Lim & Gong, 2018; Shafie et al., 2020).

The tensile strength (TS) values ranged from 6.65 ± 0.13 to 19.07 ± 1.43 MPa. When a single additive was added to the base formulation, higher TS values were observed, except for films containing CA, which showed a reduction in TS. This can be attributed to the incorporation of acetyl groups in the polymeric matrix, resulting in an expansion of free space within the films’ polymeric structure and a corresponding decrease in TS (Colussi et al., 2017). Incorporating solely OO resulted in a rise in TS accompanied by increased OO concentrations, where POO$_{1.0}$ exhibited the highest TS value. This is due to the poor plasticizing effect of OO molecules which remain confined within the vacant spaces of entanglements existing between the HMP’s chains and starch. This ultimately leads to a stronger, less flexible film, as previously stated (Shafie et al., 2020). The incorporation of further additives into the films containing OO led to a decrease in the TS values, consistent with the reduced intermolecular interactions caused by the simultaneous incorporation of two additives in the polymeric structure (Shafie et al., 2020). All films containing RH exhibited significantly high TS values, which can be attributed to the cellulose-based composition of the additive and the resulting increase in intermolecular interactions among the compounds found in the filmogenic mixtures. This can mainly be attributed to the hydrogen bonding of the OH and COOH groups of the films’ constituents (Lim & Gong, 2018).

3.6. *Comparison with non-commercial and commercial biodegradable films*

The WCA and mechanical properties of the analyzed films were evaluated and compared to commercial and non-commercial biodegradable materials commonly used in food packaging (Table 4). The commercial biodegradable films exhibited E′ values ranging from 0.25 to 4.40 GPa, TS values between 8.0 and 115.5 MPa and ETB values ranging from 2.2 to 460%. The non-commercial films presented E′ values from 0.017 to 1.27 GPa, TS between 0.028 and 16 MPa, and ETB varying from

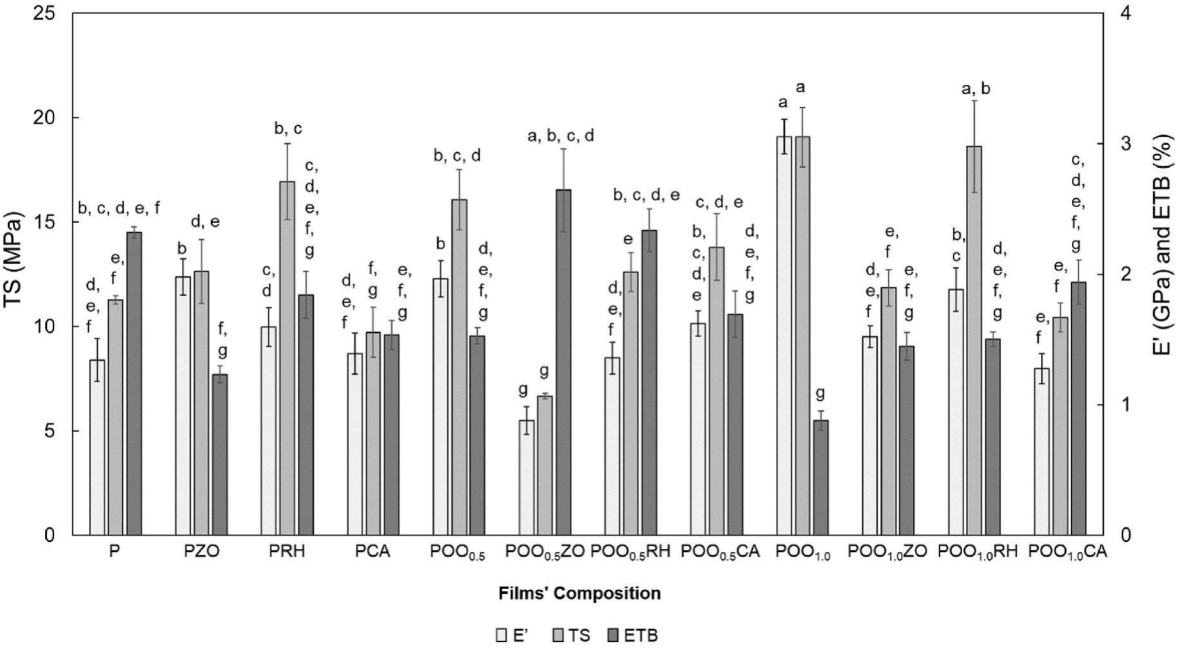

Fig. 5. Mechanical properties of the produced films. Columns with different letters differ significantly ($P < 0.05$) by Tukey test.

Table 4
Commercial and non-commercial biodegradable films' mechanical properties: elongation at break (E'), tensile strength (TS) and elongation to break (ETB); and water contact angle (WCA).

Polymers' composition	Commercial?	Applications	E' (GPa)	TS (MPa)	ETB (%)	WCA (°)	References
Solanyl® C1201 **(Polylactic acid (PLA) + Potato starch)**	Yes	Sustainable food packaging	2.0	25.0	≈2.2	–	Parres et al. (2020)
ECOVIO® F23B1 **(polybutyleneadipate-*co*-butylene terephthalate (PBAT) + PLA)**	Yes	Bags for agriculture, organic residues, fruits and vegetables, food packaging and paper coating	0.19	8.6	460	105.58	La Mantia et al. (2020)
Mater-Bi® EF04P **(Corn starch + PBAT)**	Yes	Fruits and vegetables packaging	0.13	21.1	392	111.45	
Bio-Flex® F2110 **(*co*-polyester + PLA)**	Yes	Sustainable packaging for several industries, including the food industry	0.25	8.0	95	98.6	Scaffaro et al. (2020)
Nativia® BOPLA NTSS **(PLA-based)**	Yes		–	59.84	16.70	62.5	Izdebska-Podsiadły (2021)
EarthFirst® PLA BCP **(PLA-based)**	Yes		–	27.54	6.68	72.6	
Pea starch + agar + glycerol	No	Tortillas' internal coating	0.017	0.028	186	–	Serrano et al. (2022)
Cassava starch + glycerol + soy lecithin + essential oils (lavender/fennel/lime)	No	Strawberries' packaging	0.076–1.27	1.98–4.0	16–130	56–67	Gómez-Contreras et al. (2021)
Pea starch + PLA + Pea starch modified with octenyl succinic anhydride	No		0.472	9.68	27.43	35.6	Zhou et al. (2021)
Corn starch	No	Packaging for apple slices	–	15.0	0.045	80.1	Lai and Wong (2022)
Potato starch	No		–	9.0	0.03	88.8	
Chestnut starch	No		–	16.0	0.05	79.4	

1.0 to 9.0%. The results of this study demonstrate superior E′ and TS in the films as compared to non-commercial edible films, while the ETB values remain in the same range. When compared with commercial films, those produced in this study exhibited similar ranges for E', TS and ETB, although the latter property demonstrates more distinct values based on the results of this investigation. Although certain produced films exhibited mechanical properties that could rival those found in commercial films, they possess values similar to or slightly lower than Solanyl® C1201 (Gómez-Contreras et al., 2021; Izdebska-Podsiadły, 2021; La Mantia et al., 2020; Lai & Wong, 2022; Parres et al., 2020; Scaffaro et al., 2020; Serrano et al., 2022; Zhou et al., 2021). Regarding their wettability, the films tested in this study presented contact angle values (WCA) values within the range shown in Table 4, with some of the films produced achieving higher values compared to certain commercially available films, including EarthFirst® PLA BCP or Nativia® BOPLA NTSS. Overall, the films produced in this study showed comparable or even superior mechanical properties and wettability in comparison to non-commercial and some commercial biodegradable films. These results suggest the potential for these films to compete with their counterparts in the future.

3.7. Water dissolution

To comprehend the dissolution of the produced edible films, each formulation's samples were dissolved and the turbidity of each system was assessed. These experiments were conducted at 30 °C and 60 °C, allowing for a comparison of dissolution rates under different temperature conditions, Fig. 6A and 6.B, respectively.

Fig. 6 demonstrates that, following 60 min of dissolution, the turbidity differed depending on the temperature. At 30 °C, ranged between 175 and 550 g^{-1}, whilst at 60 °C it ranged from 420 to 730 g^{-1}. This trend was expected since the rate of solid dissolution increases at higher temperatures. This is a result of more rapid movement of water molecules, the breaking of hydrogen bonds in the films' matrixes, and an increase in the films' disintegration (Li et al., 2022; Lu et al., 2022). With an increase in the films' dissolution, there is a greater abundance of

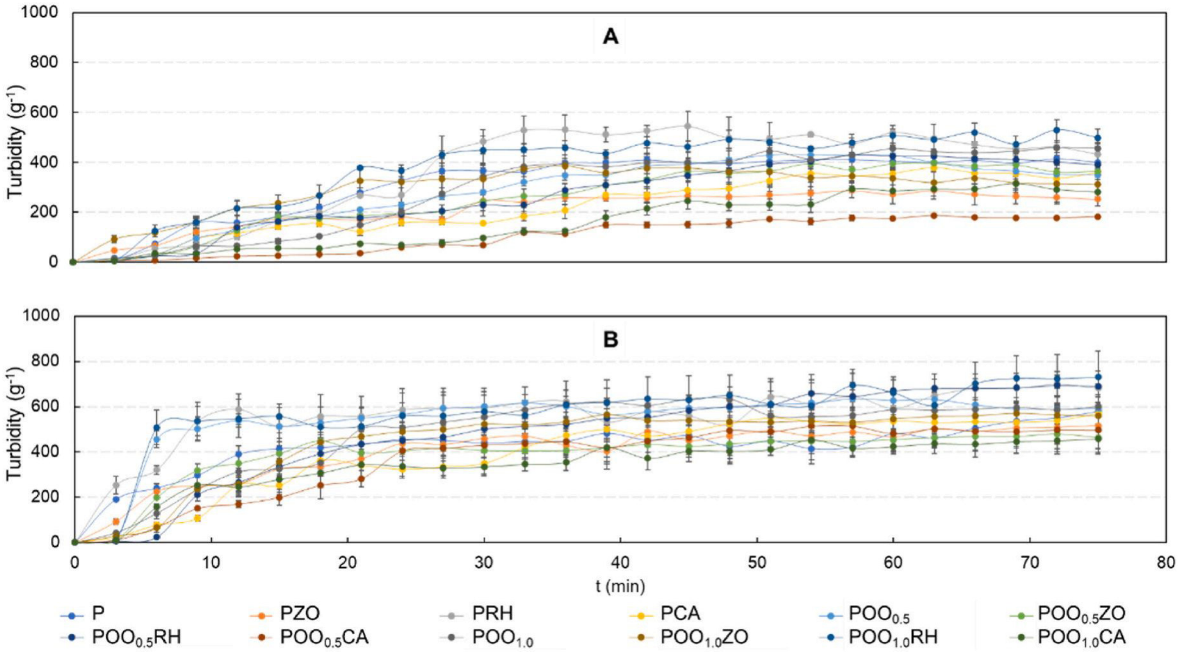

Fig. 6. The turbidity profiles for the water dissolution of all films at 30 °C (**A**) and 60 °C (**B**).

small suspended particles in the system, resulting in higher turbidity values, as observed (Serajuddin et al., 2019).

The dissolution rates of each film type were compared at two different temperatures by calculating the slopes of the turbidity curves from the data collected within 30 min. The results are presented in Table 5, displaying slopes ranging from 2.22 to 15.81 g^{-1} min^{-1} for 30 °C and 14.23 to 26.55 g^{-1} min^{-1} for 60 °C. These differences are explained by the increased dissolution phenomenon that occurs with an increase in temperature and the consequent increase in the number of suspended particles (Lu et al., 2022; Serajuddin et al., 2019). The results indicate that the CA films exhibited lower slopes at both temperatures. This suggests that the increase in turbidity caused by the dissolution of these films is smaller when compared to the other films, resulting in clear suspensions. These findings indicate that the acetylation process of the films resulted in increased free volume within the polymeric structure, thereby facilitating water penetration and solubilization (Colussi et al., 2017). Incorporation of RH into the films appears to increase the turbidity of the dissolution systems, with PRH and POO$_{1.0}$RH presenting some of the highest dissolution rates at both temperatures. The intrinsic structure of RH, which comprises lignin and silica, makes this material more impervious to water penetration and therefore has lower water solubility, which leads to a greater concentration of suspended particles in the solution system, contributing to the turbidity levels (Laftah & Wan Abdul Rahman, 2021).

4. Proof of concept

4.1. Edible hydrosoluble sachets for white sugar

After evaluation of the film properties, two formulations were selected for use as hydrosoluble sachets for white sugar (Fig. 3B). As the colour, opacity, thickness and thermal properties of all the films were very similar, these characteristics were not deemed particularly significant in selecting the most suitable films for producing the desired edible hydrosoluble sachets. One of the selected films was PCA because it has some of the lowest WVS and WVP values, the second-highest WCA, and intermediate results in terms of mechanical properties and WD. The second film, POO$_{1.0}$RH, exhibited moderate WVS and WVP, the highest TS with intermediate E' and ETB, an average WCA and one of the highest WD values. These two formulations were considered representative of the produced edible films, as they complement each other in terms of the analyzed physicochemical properties. Nevertheless, other film formulations could have been chosen as representatives for the intended purpose.

The sachets were manufactured using films from the chosen formulations and underwent analysis of weight and colour over 12 weeks. According to the results presented in Fig. 3C, the mass variance of all the sachets did not surpass 1% throughout the study (Figure S2A). The profile for both types of sachets type was similar and largely dependent on the humidity of their surrounding environment. The small observed variations of mass can be related to the humidity variations between measurement days, indicating that the sachets effectively retained their barrier properties and regulated mass transfer between the white sugar and its surroundings. These findings are noteworthy as they suggest that these films can have a broad range of uses in products that require limited heat and mass transfer to preserve their quality, including fruits and vegetables, as well as soluble coffee, tea, sugar or salt (Liu et al., 2020; Zhao et al., 2021).

Table 5
Decreasing order of turbidity slopes after 30 min of dissolution in water at 30 and 60 °C.

Film code	Slope (30 °C)	R^2	Film code	Slope (60 °C)	R^2
POO$_{0.5}$CA	2.2171	0.9754	**PCA**	14.23	0.9573
POO$_{1.0}$CA	3.2288	0.9806	**POO$_{1.0}$CA**	14.458	0.9374
PCA	6.5711	0.9478	**POO$_{0.5}$CA**	14.769	0.9916
POO$_{0.5}$RH	8.2688	0.9683	**PZO**	17.785	0.9672
PZO	8.3913	0.9565	**POO$_{0.5}$ZO**	18.384	0.9173
POO$_{0.5}$ZO	8.545	0.9765	**POO$_{0.5}$RH**	18.745	0.9785
POO$_{1.0}$	8.7498	0.9407	**P**	19.799	0.9197
POO$_{0.5}$	9.7876	0.9945	**POO$_{1.0}$ZO**	20.128	0.9809
P	13.016	0.9935	**POO$_{1.0}$**	20.679	0.9886
PRH	13.383	0.9607	**POO$_{1.0}$RH**	25.605	0.8596
POO$_{1.0}$ZO	13.525	0.9719	**POO$_{0.5}$**	26.03	0.8921
POO$_{1.0}$RH	15.808	0.9917	**PRH**	26.552	0.8923

Upon analyzing Fig. 3C, it is evident that the colour of the films remained consistent throughout all experiments, confirming the preservation of the film's aesthetic and structural properties. Additionally, the preservation is further confirmed by the findings from Figure S2B. The findings indicate that the sachets preserve both the gas and mass exchange of the packed goods and their structural integrity and appearance throughout the process, confirming that the selected films are suitable as edible hydrosoluble sachets for the preservation of white sugar. Edible films created from food by-products have demonstrated successful application as a promising material for sustainable and ecological food packaging. In addition to presenting nutritional benefits to the consumers (Chaffee & Yaros, 2007; Zhao et al., 2021), this innovation contributes to significantly reducing food waste, plastic pollution, and the environmental impact in comparison to traditional alternatives such as plastic or paper. Overall, these materials demonstrated high potential for use in the food packaging industry, as well as other sectors, as sustainable packaging.

4.2. Sensory analysis of lemon balm tea with hydrosoluble sugar sachets

Fig. 7A confirms that tea is the third food product to which respondents added sugar, corresponding to 9%, after desserts and coffee. This proof of concept aims to validate the use of hydrosoluble sugar sachets in a product where their solubilization has the potential to impact the appearance of the final product, resulting in increased turbidity of the tea.

To conduct the sensory analysis, three variations of lemon balm tea were produced. Variations A and C contained hydrosoluble sachets of PCA and POO$_{1.0}$RH, respectively, while variation B was controlled with exclusively white sugar content at the same content as the others. All three tea variations contained 10 g/L of dissolved white sugar. The tea was stored in jars and served in 30 mL cups, as illustrated in Fig. S3 (**Supplementary Data**).

A total of 60 responses were recorded for the acceptability of the produced hydrosoluble sugar sachets and 35 regarding their physical appearance. Subtle variations were noted among the samples, with the turbidity of the samples containing hydrosoluble sugar sachets being higher. However, these colour variations are not very clear in the teacups and all the teas presented acceptable colour and flavour, as confirmed by the responses from the panellists regarding the tea evaluation, Fig. S3 (**Supplementary Data**) and Fig. 7B. More than 91.7% of the participants expressed acceptance of the tea's aroma, colour, homogeneity and flavour, while 32% commented that the sugar content did not match their preferences and 75% stated that they would prefer the tea with less sugar. This implies that more than 90% of the inquired enjoy 2.5 g of sugar, or even less, per cup of tea (250 mL) (Fig. 7C). The findings suggest that increasing the water proportion in the teas can reduce the sugar concentration, thereby aligning with the preferences of the panellists. This also helps in facilitating the dissolution of sachets reducing turbidity. Additionally, the production of sachets with different sizes and quantities of sugar is an attractive option, enabling the controlled addition of the required amount of sugar in the tea and catering to the individual preferences of each consumer.

Concerning the sugar sachet's appearance, Fig. 7B shows those used for the second part of the sensory analysis. Of the 35 answers, more than 85% expressed they intend to buy hydrosoluble sugar sachets, with over 91% indicating that the appearance and colour were satisfactory. Only half of the panellists suggested that transparency in sachets is important

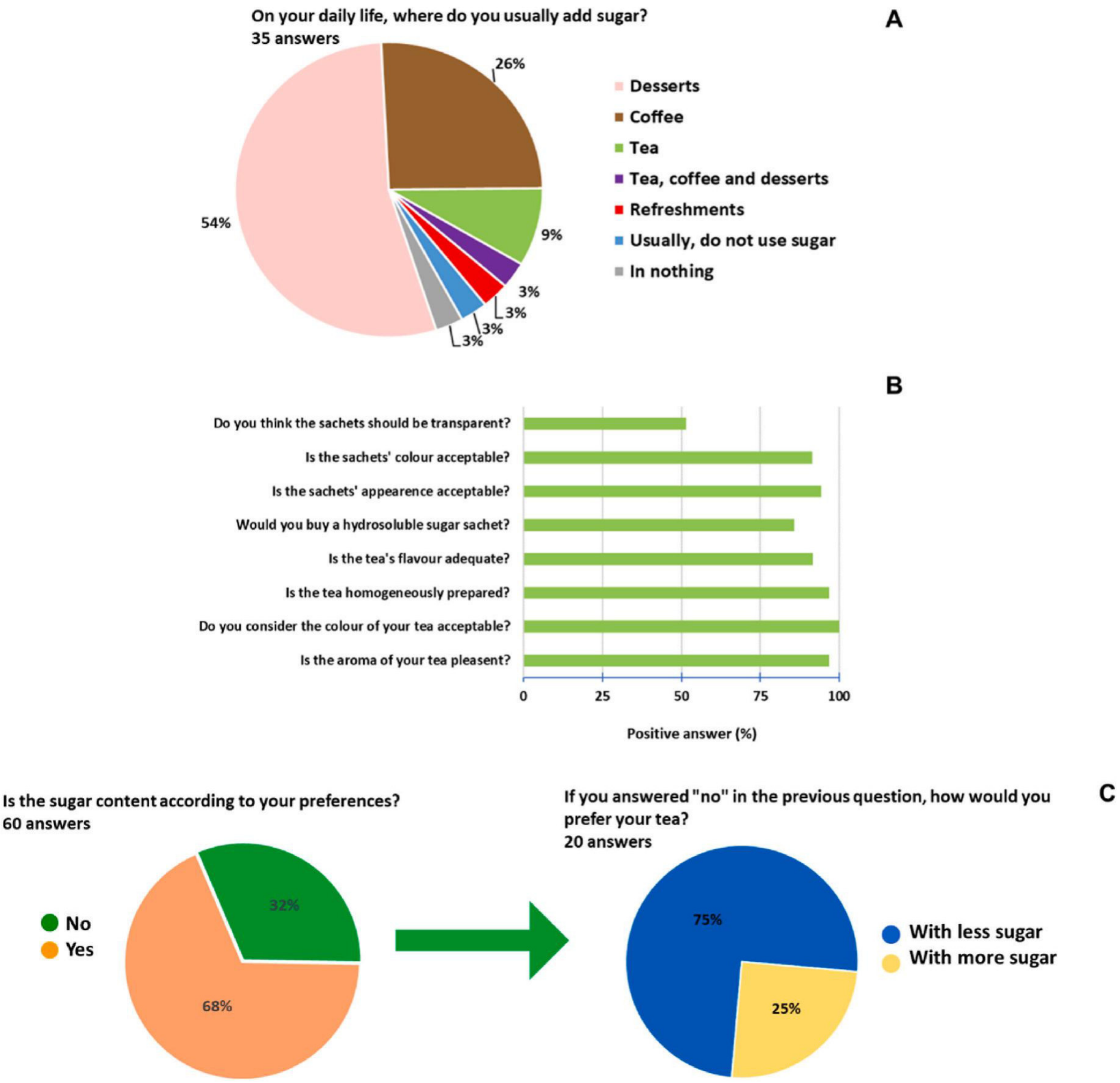

Fig. 7. Panellists’ answers for the sugar usage (**A**) and tea proof considering the hydrosoluble sugar sachets (**B**), and opinion about the sugar content (**C**).

for this pro’uct’s acceptability in the market. However, many paper sugar sachets and some plastic ones do not allow for visualization of the sugar. Only two respondents in this study reported not using sugar in their daily lives.

Thus, the implementation of edible hydrosoluble sugar sachets produced from food by-products demonstrated high acceptability, offering a promising approach to limit food waste environmental impact, as well as plastic contamination. These edible films could also be used in other applications of the packaging industry.

5. Conclusions

Orange pectin (P) played a crucial role in the properties of the films, with its high degree of esterification enhancing the mechanical resistance of the films due to the high entanglement between the pectic chains and the polymeric chains of the remaining film-forming compounds. Most of the produced films presented WCA values superior to 65°, as well as a maximum E’ value of 3.05 GPa, a maximum TS of 19.07 MPa and maximum ETB of near 3%, competing with several commercial biodegradable films in terms of wettability and mechanical properties.

The intrinsic nature and concentration of the additives were very important to the films’ physicochemical properties. Calcium acetate (CA) significantly improved the hydrophobicity, barrier properties and solubility of the edible films through polymeric chain acetylation. Films with rice husk (RH) presented enhanced mechanical and barrier properties, which was associated to the cellulosic composition of this additive and the increased number of molecular interactions with the films’ constituents. The hydrophobic nature of orange oil (OO) enhanced the films’ hydrophobicity and barrier properties, while zinc oxide (ZO) improved the hydrophilic character and ETB values of the films by increasing of the films’ surface roughness and space between the polymeric chains.

All films underwent complete dissolution after 15 min in water at 60 °C, with PCA and POO$_{1.0}$RH being identified as the most representative among the analyzed films. The selected films were used to produce hydrosoluble sachets for white sugar, which retained their structural integrity throughout a 12-week storage period and presented a high level of acceptability in a sensory analysis using lemon balm tea sugared with these sachets (over 90% of the respondent satisfied with the consumed tea and more than 85% interested in purchasing and consuming the sachets). Due to the good mechanical and barrier properties, enhanced dissolution and acceptability by the panelists, POO$_{1.0}$RH was selected as the best film produced in this study.

Overall, hydrosoluble sachets made from edible films produced using agrifood residues and additives were successfully utilized for the preservation of white sugar. The study revealed that this kind of film could be considered a promising solution for sustainable food packaging, along with other eco-friendly applications within the packaging industry.

CRediT authorship contribution statement

Alexandre M.S. Jorge: Writing – original draft, Visualization, Validation, Software, Investigation, Formal analysis, Data curation. **Cristiana S.A. Bento:** Writing – review & editing, Validation, Investigation, Data curation. **Marta H.F. Henriques:** Writing – review & editing, Resources, Investigation. **Marisa C. Gaspar:** Writing – review & editing, Visualization, Validation, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization. **Mara E.M. Braga:** Writing – review & editing, Visualization, Validation, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodhyd.2024.109776.

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