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Tailoring the surface properties and flexibility of starch-based films using oil and waxes recovered from potato chips byproducts

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Abstract

Agrofood byproducts may be exploited as a source of biomolecules suitable for developing bioplastic materials. In this work, the feasibility of using starch, oil, and waxes recovered from potato chips byproducts for films production was studied. The recovered potato starch-rich fraction (RPS) contained an amylopectin/amylose ratio of 2.3, gelatinization temperatures varying from 59 to 71 °C, and a gelatinization enthalpy of 12.5 J/g, similarly to a commercial potato starch (CPS). Despite of its spherical and oval granules identical to CPS, RPS had a more amorphous structure and gave rise to low viscous suspensions, contradicting the typical B-type polymorph crystal structure and sluggish dispersions of CPS, respectively. When used for films production, RPS originated transparent films with lower roughness and wettability than CPS-based films, but with higher stretchability. In turn, when combined with RPS and CPS, oil or waxes recovered from frying residues and potato peels, respectively, allowed to develop transparent yellowish RPS and CPS-based films with increased surface hydrophobicity, mechanical traction resistance, elasticity, and/or plasticity. Therefore, potato chips industry byproducts revealed to have thermoplastic and hydrophobic biomolecules that can be used to efficiently develop biobased plastics with improved surface properties and flexibility, opening an opportunity for their valorization.

Keywords. Bioplastics; Hydrophobicity; Plasticity

1. Introduction

According to Food and Agricultural Organization (FAO), more than 320 million tonnes of potatoes are being annually cultivated worldwide on 20 million hectares of land, ranking potato in the fourth position amongst the worlds' most important food crops. From those, around 15% are mainly processed for chips [1]. Nevertheless, a change in eating habits, as well as in the lifestyle, raised the demand for potato chips consumption, and the potato chips industry business is expected to grow. Consequently, potato chips byproducts should increase, namely the non-edible washing slurries, potato peels, and frying residues, which are very rich in starch, waxes, and oil, respectively.

Starch is a mixture of two glucose polymers, amylose and amylopectin, linked through α-D-(1 → 4) glycosidic bonds with low or high amount of α-D-(1 → 6) glycosidic branches, respectively [2]. These polysaccharides can be extracted from different botanical sources of plants, allowing to obtain starch with different physicochemical properties [3-5]. Within potato starch, even when obtained from different tuber varieties, a constancy of physicochemical properties has been reported [6, 7]. Independently of its source, starch, in the presence of plasticizers and by the action of heat and shear stress, has film-forming ability. This phenomenon occurs due to the starch granules destruction, allowing the short-branched chains in amylopectin to gelatinize, giving rise to thermoplastic starch [8]. However, the inherent hydrophilicity and high density of starch (1400 kg/m^3) leads to quite hygroscopic and brittle materials [9], thus limiting their application range. To enhance these properties, starch has been blended with plasticizing agents as glycerol, xylitol, sorbitol, and ascorbic acid [10-12], and synthetic polymers as polypropylene [13], polyethylene [14], polycaprolactone [15], and polylactic acid [16, 17]. Nevertheless, these approaches increase the materials commercial cost, a key factor in the competitive bioplastics market, and compromise the pristine biodegradability of starch.

In this work it is hypothesized that starch recovered from potato washing slurries can be used for developing biobased plastics whose properties can be modulated by the incorporation of waxes and oil also recovered from potato chips industry byproducts. For comparison purposes, a commercially available potato starch was used as reference.

2. Materials and Methods

2.1. Materials

Potato chips byproducts (washing slurries, potato peels, and frying residues) were supplied by A Saloinha, Lda., a Portuguese potato chips company. Commercial potato starch was provided from Sigma Aldrich (Germany) as well as the glucose (hexokinase) assay kit. Glycerol was supplied from Scharlab S. L. (Spain). Chloroform and *n*-hexane were obtained from Carlo Erba Reagents S.A.S. (Italy). All purchased reagents were of analytical grade and used without further purification.

2.2. Recovery of starch from potato washing slurries

Potato washing slurries were pre-weighed, frozen, and freeze-dried. To separate starch from small potato slices remaining in the potato washing slurries, the freeze-dried material was sieved using a stainless-steel sieve with a 125 μm mesh. The recovered starch was stored in a desiccator containing silica gel until further use. The recovery yield of starch (g of starch per 100 g of lyophilized potato washing slurries) was determined.

2.3. Starch characterization

2.3.1. Starch, amylose, and amylopectin content

Starch content was determined by amyloglucosidase hydrolysis, using the hexokinase assay for glucose quantification [18]. Prior to the starch samples disintegration and dissolution, soluble sugars were removed by dispersing 200 mg of starch in 10 mL of ethanol (40% V/V), stirred, centrifuged (3000 rpm), and the supernatant discarded. This extraction was one time repeated. Afterwards, the sample was suspended in 10 mL of DMSO and heated at 100 ºC during 30 min under stirring, allowing starch granules disintegration. After cooling down, 1.7 mL of 8 M HCl was added and the mixture was stirred during 30 min at 60 ºC for complete starch solubilization. The solution was neutralized with 5 mL of 4 M NaOH and 2.5 mL of 2.5 M sodium acetate solution, adjusting the pH to 4.8 with 1 M NaOH and performing a total volume of 100 mL with distilled water. To 5 mL of sample solution, 0.125 mL of amyloglucosidase solution (prepared in 0.2 M acetate buffer, pH 4.8) was added and the enzymatic reaction was carried out at 60 ºC overnight. After enzyme inactivation in a boiling water bath, the solutions were cooled down and mixed with Carrez I solution (0.125 mL). This last step was repeated with the same volume of Carrez II solution. After centrifugation (3000 rpm), glucose quantification was performed using the Glucose (HK) assay kit. To 75 µL of the dissolved starch solution, 1 mL of Glucose (HK) Assay Reagent was added, and the enzymatic reaction was carried out at room temperature for 15 min. The starch conversion into glucose was monitored by measuring the absorbance at 340 nm. For the sample blank preparation, the Glucose (HK) Assay Reagent volume was replaced by distilled water. In turn, for Glucose (HK) Assay Reagent blank, the starch solution volume was also substituted by distilled water. A glucose solution (1 mg/mL) was used as standard. The glucose amount was calculated following equations 1 and 2. This assay was carried out in triplicate and the glucose concentrations obtained were averaged.

(Eq. 1) $$A_{540}\ (total\ blank) = A_{540}\ (sample\ blank) + A_{540}\ (reagent\ blank)$$

(Eq. 2) $$\text{mg glucose/mL} = \frac{\Delta A_{540}\ (TV)(glucose\ Mw)(F)}{(\varepsilon)(d)(SV)(Conversion\ factor\ for\ \mu g\ to\ mg)}$$

where: ΔA_{540} = A_{540} test – A_{540} total blank; TV is the total volume (mL); F is the dilution factor; ε is the millimolar extinction coefficient for NADH at 340 nm (mL/μmol.cm^{-1}); d is the light path (cm); and SV is the sample volume (mL).

Amylose and amylopectin content of potato starch samples was assessed by gel permeation chromatography (GPC) using a starch dispersion (1% w/V) pre-dissolved in boiling water. The SEC analysis was carried out using two PL aquagel-OH MIXED 8 μm columns (300 x 7.5 mm) protected by a PL aquagel-OH Guard 8 μm pre-column on a PL-Gel Permeation Chromatograph 110 system (Polymer Laboratories, UK). All the systems were maintained at 36 ºC during analysis. Standards and samples were dissolved in 0.1 M $NaNO_3$ aqueous solutions to a concentration of 5 mg/mL. The eluent (0.1 M $NaNO_3$ solution) was pumped at a flow rate of 0.9 mL/min. The columns were calibrated with pullulan standards (Polymer Laboratories, UK) in the range of 5.8 – 380 kDa.

2.3.2. Gelatinization and pasting properties

Gelatinization temperatures and enthalpy changes of potato starch were determined by differential scanning calorimetry (DSC) using a PYRIS Diamond equipment. Starch dispersions (30% w/V) were prepared in distilled water, transferred into stainless steel crucibles of 24 bar maximum pressure, sealed, and heated from 25 to 100 ºC with a heating rate of 10 ºC/min. The instrument was calibrated towards indium and plumb using an empty crucible as reference. The onset (T_o), peak (T_p), and conclusion (T_c) temperatures, and gelatinization enthalpy (ΔH) were directly obtained from DSC thermograms.

The pasting properties of potato starch were determined using a Rapid Visco-Analyzer (Perten RVA 4500, Australia). Starch dispersions (5% w/V) pre-equilibrated (25 °C for 1 min) were heated from 25 to 95 °C at 14 °C/min with stirring speed of 160 rpm and, after maintaining the temperature at 95 °C for 3 min, cooled down to 25 °C, following the same temperature rate [19].

2.3.3. Starch granules shape, size, and crystallinity

The potato starch granules were observed using a high-resolution field emission scanning electron microscope (Hitachi SU-70). Samples were coated under an argon atmosphere with a thin layer of gold-palladium at 10 kV and 10 mA for 3 min using a Polaron sputter coater E5000.

Potato starch granules size distribution was determined by light scattering using a Coulter counter LS 230 with a detection limit of 0.04 – 2000 µm. The system follows a Fraunhofer optical model. Each starch sample was previously dispersed in distilled water and ultra-sonicated for a few seconds to avoid granules' agglomeration. Measurements were carried out in triplicate.

X-ray diffractograms of potato starch dried and preconditioned at 30, 40, 80, and 98% of relative humidity were obtained using a PANalytical Empyrean X-ray diffractometer. The radiation used was Cu-Kα (wavelength of 0.15406 nm). The scan was carried out at 45 kV and 40 mA for 2θ range of 4 - 40º with a step size of 0.04º and a collection time of 196 s at each step.

2.4. Recovery and characterization of oil from potato frying residues

The lipidic fraction was extracted from 40 g of frying residues using a Soxhlet extractor at 80 ºC, for 5 h, with 300 mL of *n*-hexane. After removing the residual water content with anhydrous sodium phosphate, the solvent was evaporated under reduced pressure until constant weight. The recovery yield of oil (g of lipid per 100 g of frying residues) was determined in triplicate and averaged.

Esterified fatty acids were analyzed as fatty acid methyl esters (FAME) after transesterification of recovered lipidic fraction [20]. After adding cooled 0.8 mL of heptadecanoate methyl ester (1.5 g/L prepared in *n*-hexane) and 0.2 mL of a KOH methanolic solution (2 M) to 10 mg of lipidic extract, the mixture was vigorously shaken. Then, 2 mL of saturated NaCl solution was added and, after centrifuging at 2000 rpm during 5 min, 0.5 mL of the organic phase was collected and analyzed in a GC-FID PerkinElmer-Clarus 400 with a capillary column DB-FFAP (30 m x 0.32

mm x 0.25 µm). The oven program followed 3 ramps of temperatures: (i) 75 ºC to 155 ºC at a rate of 15 ºC/min; (ii) increase to 180 ºC at a rate of 3 ºC/min; and (iii) increase to 220 ºC at a rate of 20 ºC/min (3 min). Injector and detector temperatures were 245 ºC and 250 ºC, respectively. Hydrogen was used as carrier gas.

2.5. Recovery and characterization of waxes from potato peels

Waxes were recovered from potato peels through a solid-liquid extraction at 37 ºC during 24 h using chloroform as solvent. After filtration, the solvent was evaporated under reduced pressure [21]. The recovery yield of waxes (g of waxes per 100 g of lyophilized potato peels) was determined in triplicate and averaged.

ATR-FTIR analysis was used to confirm that only the lipidic fraction was recovered during waxes extraction process. The FT-IR profile of recovered waxes was obtained using a Golden Gate single reflection diamond ATR system in a Perkin Elmer Spectrum BX spectrometer. Spectra were recorded at the absorbance mode from 4000 to 500 cm^{-1} at a resolution of 16 cm^{-1} through 64 co-added scans.

2.6. Films production

Starch was suspended in distilled water (4% w/V) and further gelatinized at 95 ºC during 30 min under stirring. After filtration under vacuum through a porous glass filter (G1), glycerol (30% w/w of dry starch weight) was added. Posteriorly, oil or waxes (1; 2; and 3% w/w of dry starch weight) were added into the gelatinized starch suspension at 95 ºC, stirred during 15 min, and further homogenized using a

Micro D9 disperser at 26,000 rpm during 2 min plus 19,000 rpm during 3 min [22]. The mixture was then degassed, transferred onto plexiglass plates of 144 cm^2 with 3 mm deep, and the solvent was overnight evaporated in an air circulating oven at 25 ºC. Starch films without oil or waxes were used as control. Commercial potato starch was used as reference. The films were stabilized at room temperature under controlled relative humidity (54% RH) for at least 5 days until further characterization.

2.7. Films characterization

2.7.1. Optical properties

The color parameters of each film were determined, in triplicate, using a portable Chroma Meter (Minolta CR-400 Japan). CIELab color profile was used to express the L^* (luminosity), *a** (red/green color), and *b** (yellow/blue color) components. The total difference in color (ΔE) was calculated following equation 3.

(Eq. 3)
$$\Delta \mathrm{E} = \sqrt{(\Delta L^{*})^2 + (\Delta a^{*})^2 + (\Delta b^{*})^2)}$$

where: ΔL^*, Δa^*, and Δb^* are the L^*, a^*, and b^* differences between films containing oil or waxes and the control films [23, 24].

2.7.2. Topography

The topography of each film was evaluated by atomic force microscopy (AFM) with a multimode Nanoscope III microscope (Digital Instruments). Scans were performed in tapping mode using a standard silicon AFM probe (µmasch, HQ:NSC14/Al BS), with a resonance frequency of 160 kHz and a constant force of 5.0 N/m. The open-source software Gwyddion version 2.47 was used to generate the two- and three-dimensional images and to calculate the highest peak height and mean surface roughness (*Ra*), *i.e.* the arithmetic average of absolute profile heights' values over the length evaluation.

2.7.3. Wettability

Static water contact angles were used to determine the wetting properties of each film surface using a Dataphysics OCA instrument, fitted with an automatic image capture system (Dataphysics SCA20 M4). Three strips (60 mm x 10 mm) were cut in distinct areas of the films. A 3 µL drop of ultrapure water was dispensed onto each strip surface using a microsyringe until proper contact between the drop and strip surface was made. Drop shape analysis was used to determine the contact angle by the sessile drop fitting method. When the contact time between the water drop and the film was no greater than 30 s, pictures were taken and the contact angle values were obtained following the Young-Laplace equation [25, 26]. At least 10 measurements on different areas of the strip, each equivalent to a drop, were averaged.

2.7.4. Solubility in aqueous medium

The films solubility in aqueous medium was measured as dry matter percentage that solubilized in an aqueous medium for 7 days. Squares (4 cm^2) of each film were cut, weighed, immersed in distilled water (pH 6.5) containing sodium azide, and stirred at room temperature with orbital agitation (80 rpm). Nonsolubilized samples were carefully separated using a spatula, dried at 105 °C for 16 h, cooled down in a desiccator containing silica gel, and further weighed. Residual humidity of each film was determined by drying at 105 °C during 16 h, cooling down, and weighing. The solubility and residual humidity of each film were calculated by weight loss percentage in triplicate and averaged.

2.7.5. Mechanical properties

The films traction resistance, elasticity, and plasticity were determined following the ascribed ASTM D 882-83 standard method using a texture analyzer apparatus (model TA.Hdi, Stable Micro Systems) equipped with fixed grips lined with thin rubber on the ends. The initial set grip separation was set at 50 mm and the crosshead speed was set at a constant rate of 0.5 mm/s [27]. Each film was cut in 12 strips of 90 mm length and 10 mm width and used to determine the mechanical profile of each formulation. Tensile strength, Young's modulus, and elongation at break values were calculated following equations 4, 5, and 6.

(Eq. 4)

$$\text{Tensile strength (MPa)} = \frac{load\ at\ break}{(original\ width) * (original\ thickness)}$$

(Eq. 5)

$$\text{Young's modulus (MPa)} = \frac{\frac{(load\ at\ point\ on\ tangent)}{(original\ width) * (original\ thickness)}}{\frac{(elongation\ at\ point\ on\ tangent)}{(initial\ gage\ length)}}$$

(Eq. 6)

$$\text{Elongation at break (\%)} = \frac{(elongation\ at\ rupture)}{(initial\ gage\ length)} * 100$$

2.8. Statistical analysis

All the obtained data were statistically analyzed through one-way and two-way ANOVA tests. GraphPad Prism 7.0 for Windows was the software used. Significant differences were identified as $p < 0.05$.

3. Results and discussion

Freeze-dried potato washing slurries was composed by 44% of solid matter containing starch and small potato slices. Aiming to separate starch from the small potato slices, the solid fraction was sieved using a stainless sieve with 150 µm mesh. As a result, 76% of starch, related to dry solid matter weight, was recovered. Therefore, *per* each kg of potato washing slurries was possible to obtain 334 g of recovered potato starch-rich fraction (RPS).

3.1. Characterization of RPS *vs* CPS

Table 1 shows the starch content, amylopectin/amylose ratio, and gelatinization properties of RPS and CPS.

Table 1: Starch content, amylopectin/amylose ratio, and gelatinization properties (onset (T_o), peak (T_p), and conclusion (T_c) temperatures, and gelatinization enthalpy (ΔH)) of recovered potato starch-rich fraction (RPS) and commercial potato starch (CPS)

	RPS	**CPS**
Starch content (% in dry matter)	72.0 ± 0.8	72.2 ± 1.2
Amylopectin/Amylose ratio	2.3	2.7
Gelatinization properties		
T_o (ºC)	55.1 ± 1.4	58.2 ± 0.8
T_p (ºC)	59.0 ± 0.1	62.0 ± 1.1
T_c (ºC)	71.4 ± 0.1	74.4 ± 0.1
$T_c - T_o$ (ºC)	16.4 ± 1.5	16.2 ± 0.8
ΔH (J/g, db)	11.8 ± 3.6	12.7 ± 1.3

RPS showed a starch content identical to CPS (*ca.* 72%) and contained two distinct molecular weight populations, as identified by two GPC chromatogram peaks (**Fig. S1**). The most abundant population presented the highest molecular weight (350 kDa), while the second one had a molecular weight of around 50 kDa. In starch, the highest molecular weight polymer is attributed to amylopectin [28]. Therefore, the first peak was related with the amylopectin fraction, while the second one referred to the amylose portion. GPC analysis also allowed to verify that RPS and CPS showed an amylopectin/amylose ratio of 2.3 and 2.7, respectively (**Table 1**), which corresponded to approximately 70% of amylopectin and 30% of amylose in RPS and 73% of amylopectin and 27% of amylose in CPS. The amylose content allows to categorize starch as 'waxy' (< 15%), 'normal' (20 - 35%) or 'high' (> 40%) [29]. Therefore, RPS and CPS used in this work were both 'normal starches'.

Regarding the gelatinization properties, both RPS and CPS evidenced well-defined single DSC endotherms (**Fig. S2**), where the gelatinization onset (T_o), peak (T_p), and conclusion (T_c) temperatures ranged from 55.1 ºC to 58.2 ºC, 59.0 ºC to 62.0 ºC, and 71.4 ºC to 74.4 ºC, respectively (**Table 1**). Therefore, RPS showed T_o, T_p, and T_c values slightly lower than CPS. Moreover, RPS and CPS presented quite similar gelatinisation temperature ranges (T_c - T_o), around 16.4 ºC and 16.2 ºC, respectively. These results are comparable to values previously reported for native potato starch in which T_o, T_p, and T_c range from 58.7 ºC to 62.5 ºC, 62.5 ºC to 66.1 ºC, and 68.7 ºC to 72.3 ºC, respectively [6, 7, 30]. Furthermore, RPS and CPS showed gelatinization enthalpies (ΔH) of 11.8 J/g and 12.7 J/g, respectively, fitting with the ΔH values reported for potato starch that ranged from

12 to 24 J/g [30, 31]. Starch gelatinization enthalpy gives an overall measure of molecular organization within the starch crystalline regions related to the distribution and association of amylopectin chains. This parameter is an indicator of the molecular order loss due to hydrogen bond breaking within the starch granules that occur during heating [7, 30, 32]. Thus, it can be considered that RPS had a slightly less organized molecular structure than CPS, possibly caused by the industrial processing that this starch sample suffered. Nevertheless, the lowest enthalpy value observed for RPS may suggest that a lower activation energy will be required to initiate its gelatinization, an important advantage for the RPS use on films production.

Concerning the morphological properties, **Fig. 1** shows that both RPS and CPS had spherical and oval granules (**Fig. 1A**) with an average size ranging from 15.4 µm to 62.7 µm for RPS and from 14.9 µm to 69.8 µm for CPS **(Fig. 1B)**. These characteristics corroborate with the smooth-surfaced, oval and irregular shape, and size described for native potato starch granules (diameter < 110 µm) [6, 33]. **Fig. 1C** shows the X-ray diffraction (XRD) patterns of RPS and CPS determined at 98% of relative humidity. The pre-conditioning moisture was selected based on the highest crystallinity profile achieved (**Fig. S3**). XRD diffractograms of RPS showed reflections at 5.6º, 15.0º, 17.2º, 22.4º, and 24.1º 2θ, comparable with the XRD pattern of CPS, although less intense, assigned to the B-type polymorph crystal structure typical of native potato starch [34]. These diffractogram changes revealed that the industrial processing affected the RPS molecular organization, corroborating with its slightly low gelatinization enthalpy (**Table 1**).

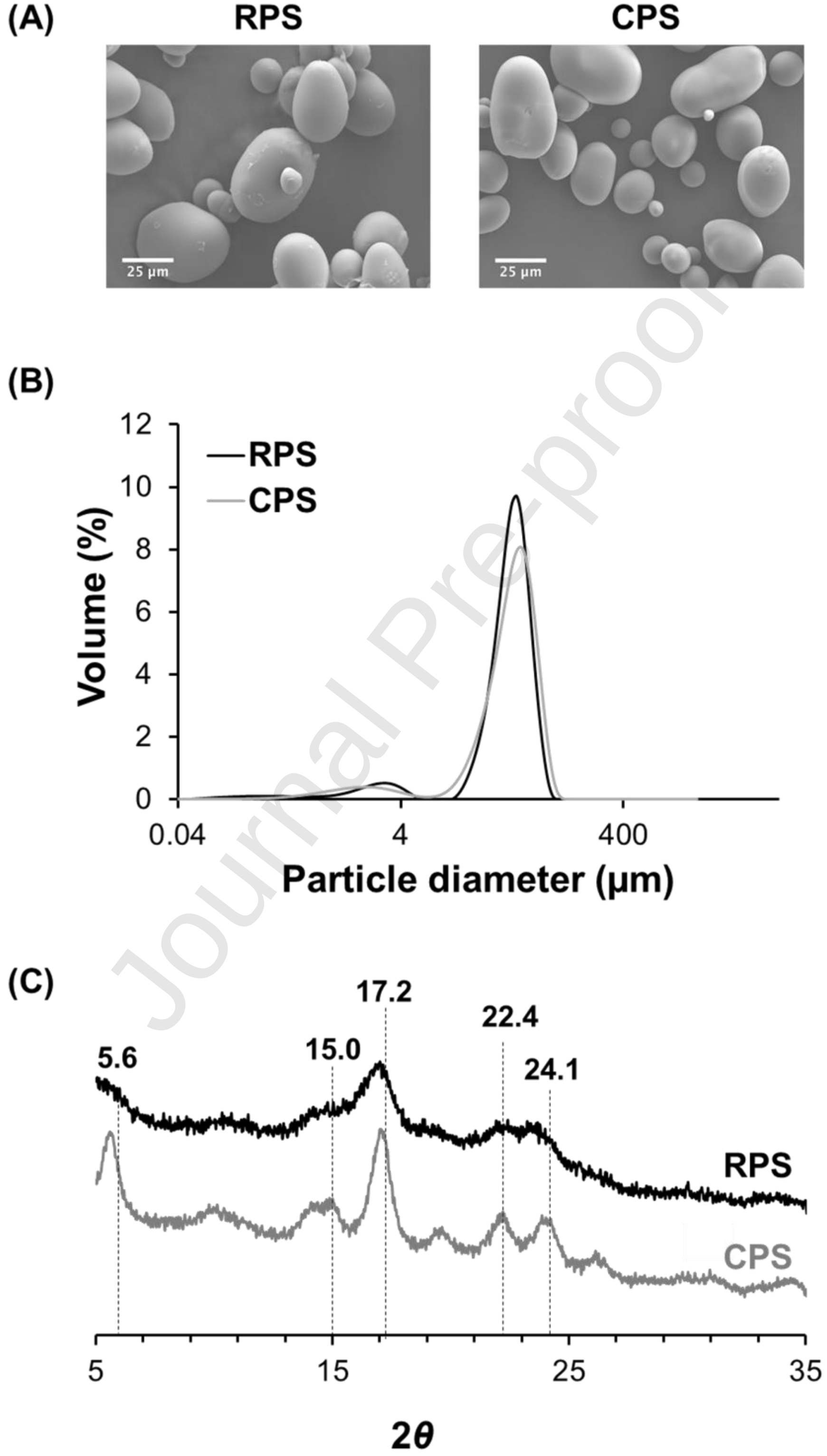

Fig. 1: Recovered potato starch-rich fraction (**RPS**) and commercial potato starch (**CPS**) granules shape, size, and crystallinity. **(A)** Scanning electron microscopy micrographs, **(B)** size distribution, and **(C)** XRD pattern.

For complementing the information of starch granules shape, size, and crystallinity, the pasting profile of both RPS and CPS was determined and compared. At around 90 ºC, RPS showed a viscosity peak of 612 mPa.s (**Fig. 2**), 4 times less than the viscosity value observed for CPS (2522 mPa.s). This phenomenon may be related with the slightly higher amylose content in RPS, as indicated by the amylopectin/amylose ratio (**Table 1**), thus decreasing the RPS water sensitivity and thermomechanical processability during gelatinization [35].

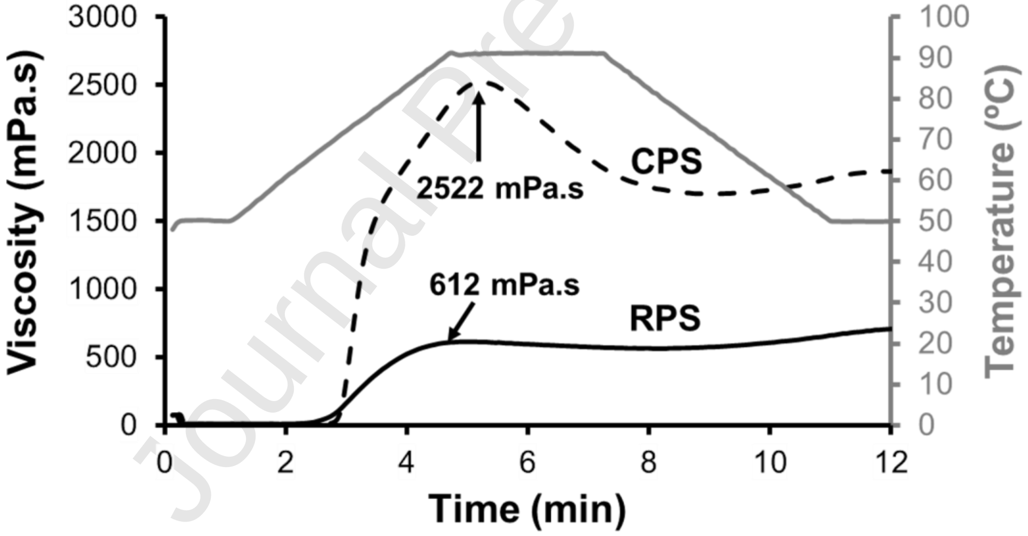

Fig. 2: Pasting profiles of recovered potato starch-rich fraction (**RPS**) and commercial potato starch (**CPS**) suspensions (5% w/V) prepared in distilled water.

3.2. Characterization of potato starch/lipids-based films

Targeting to overcome the hygroscopic and brittleness of starch-based films, the influence of oil and waxes recovered from potato frying residues and potato peels, respectively, on physicochemical and mechanical properties of RPS- and

CPS-based films was studied. Herein, in a first instance, the lipidic fractions were recovered. From potato frying residues, 61% (w/w) of oil was extracted, having the following composition in fatty acids: 43% oleic, 35% palmitic, 14% linoleic, 3% stearic, 1% lauric, 0.5% capric, 0.3% linolenic, 0.3% palmitoleic, and 0.2% myristic acid residues (**Fig. S4**). On the other hand, from potato peels were extracted 0.5% (w/w) of waxes, whose lipidic profile was confirmed by FTIR (**Fig. S5**), showing the typical triacylglyceride bands at 3600 – 3100 cm^{-1} (OH stretching), 2918 – 2848 cm^{-1} (aliphatic C-H stretching), and 1739 – 1700 cm^{-1} (C=O stretching) [21].

3.2.1. Optical properties

Table 2 shows the chromatic parameters and total color differences among RPS- and CPS-based films with ad without lipidic molecules. When used for films production, RPS allowed to attain materials with an average lightness (*L** value) of 91.55, statistically similar to the CPS-based films (*L** = 92.98). The incorporation of oil or waxes did not significantly change the *L** value of RPS-based films. This behavior was also observed on CPS-based formulations, except for 2% or 3% of waxes that significantly decreased the CPS-based films *L** value. Moreover, both *a** (red-green) and *b** (yellow-blue) coordinates of RPS and CPS-based films significantly changed, although in small proportion, with the incorporation of oil or waxes. For RPS-based films, *a** value decreased from 0.87 up to 0.59 with oil incorporation, while *b** value increased from -1.85 to -0.93, according to the oil amount used. RPS/waxes-based films showed the same *a** decrease (up to 0.45) and *b** increase (up to -0.22) tendency. On the other hand, oil doping decreased the CPS-based formulations *a** value up to 0.17, while maintaining their *b** value in around -0.20, and waxes mixture decreased the CPS-based films *a** value up to 0.04 and significantly increased their *b** value up to 1.40.

Table 2: Mean values for lightness (*L*),* red-green *(a*),* yellow-blue *(b*),* and total color (ΔE) of recovered potato starch-rich fraction (RPS)- and commercial potato starch (CPS)-based films, containing different amounts (0; 1; 2; and, 3% (w/w) of dry starch weight) of oil (O) or waxes (W). Different letters represent significantly different values ($p < 0.05$)

		*L**	*a**	*b**	ΔE*
RPS	**Control**	91.55 ± 0.36^{a}	0.87 ± 0.04^{b}	-1.85 ± 0.04^{i}	---
	O1%	91.61 ± 0.35^{a}	0.73 ± 0.02^{c}	-1.89 ± 0.02^{i}	0.16
	O2%	90.97 ± 0.37^{a}	0.60 ± 0.06^{d}	-1.08 ± 0.02^{j}	1.00
	O3%	91.14 ± 0.28^{a}	0.59 ± 0.03^{e}	-0.93 ± 0.08^{k}	1.04
	W1%	91.92 ± 0.16^{a}	$0.72 \pm 0.02^{f,c}$	-1.39 ± 0.05^{l}	0.61
	W2%	91.31 ± 0.53^{a}	$0.57 \pm 0.01^{g,d}$	$-0.94 \pm 0.04^{m,j}$	0.99
	W3%	91.26 ± 0.32^{a}	0.45 ± 0.04^{h}	-0.22 ± 0.06^{n}	1.71
CPS	**Control**	92.98 ± 0.15^{o}	0.58 ± 0.02^{r}	-0.21 ± 0.02^{y}	
	O1%	93.02 ± 0.12^{o}	0.29 ± 0.02^{s}	-0.21 ± 0.08^{y}	0.29
	O2%	92.94 ± 0.30^{o}	0.21 ± 0.03^{t}	-0.21 ± 0.07^{y}	0.37
	O3%	93.01 ± 0.43^{o}	0.17 ± 0.05^{u}	-0.20 ± 0.05^{y}	0.41
	W1%	92.88 ± 0.14^{o}	0.40 ± 0.02^{v}	0.21 ± 0.11^{z}	0.47
	W2%	92.34 ± 0.23^{p}	$0.22 \pm 0.02^{w,t}$	0.88 ± 0.13^{A}	1.31
	W3%	92.17 ± 0.14^{q}	0.04 ± 0.05^{x}	1.40 ± 0.34^{B}	1.88

Overall, both oil or waxes promoted a yellowish coloration to RPS- and CPS-based films, whose total color difference (ΔE) was directly related to the lipid amount. Nevertheless, the inherent transparency of RPS- and CPS-based films was not compromised by the oil and waxes incorporation, as observed on the films real images (**Fig. S6**). These optical changes are in accordance with reported data in which ΔE value is significantly affected by essential oils and beeswax, candelilla or carnauba waxes [24, 36].

3.2.2. Surface topography

Fig. 3 shows the topographical changes promoted by oil or waxes on both top and bottom surfaces of RPS- and CPS-based films.

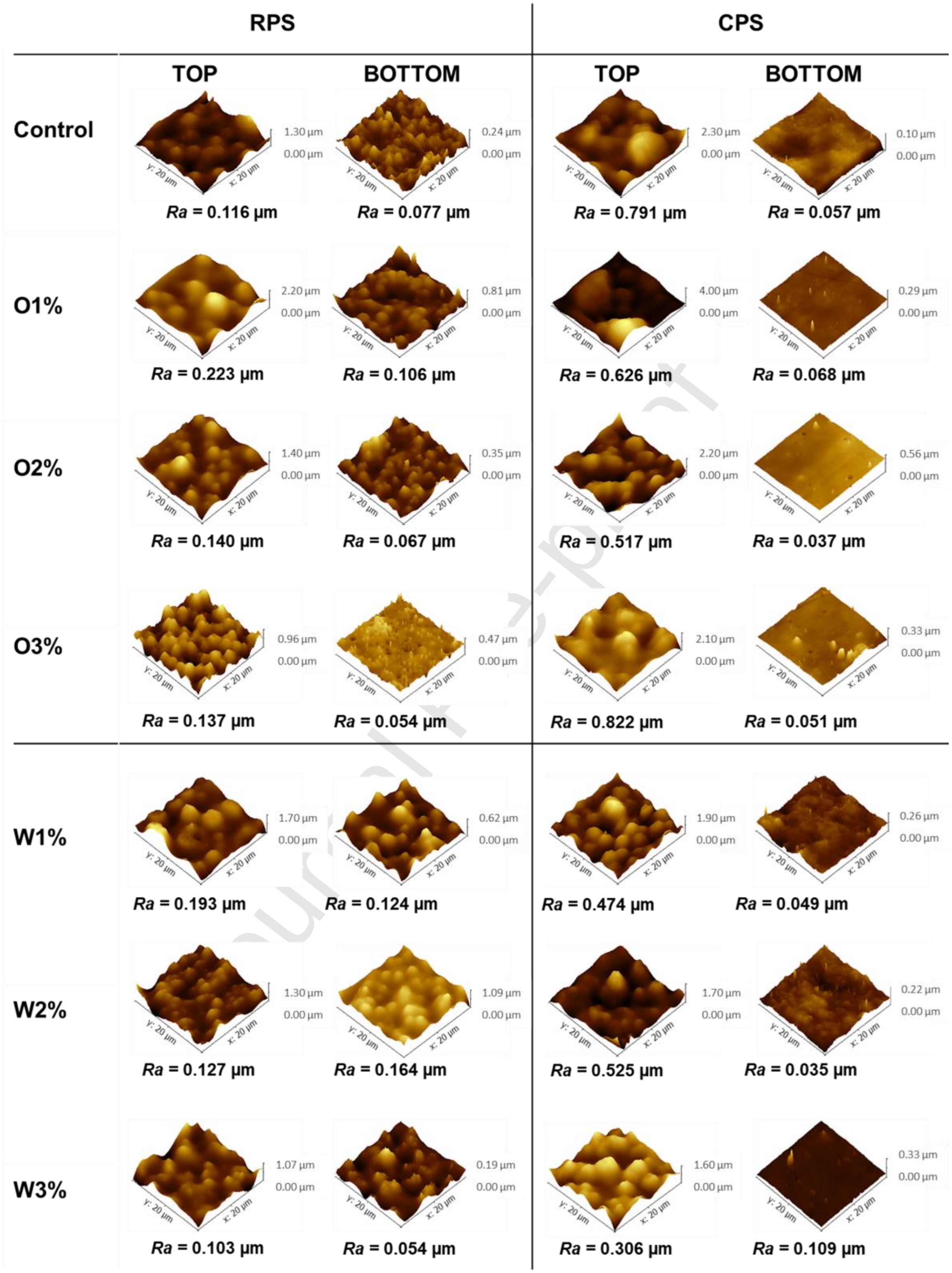

Fig. 3: 3D AFM topographic images of recovered potato starch-rich fraction (**RPS**) and commercial potato starch (**CPS**)-based films, containing different amounts (0; 1; 2; and, 3% (w/w) of dry starch weight) of oil (**O**) and waxes (**W**). **TOP** and **BOTTOM** are related with the film faces exposed to air and in direct contact with the plexiglass plate during the solvent casting process, respectively.

RPS-based films evidenced a hill-valley-structure, having a top surface containing peaks with a maximum height of 1.30 μm and a roughness (*Ra*) value of 0.116 μm, and a bottom surface with peaks having a maximum height of 0.24 μm and a *Ra* value of 0.077 μm. Therefore, RPS-based films evidenced a top surface rougher than the bottom one. This profile was also present in CPS-based films, but with a more pronounced roughness discrepancy between both surfaces than the RPS-based films. On the other hand, the incorporation of oil or waxes increased the peak height and *Ra* on both top and bottom surfaces of RPS-based films, having an opposite effect on CPS-based films. Furthermore, the highest films roughness was promoted by oil incorporation.

Solvent casting technique is known to be challenging on creating homogenous materials, particularly when a mixture of compounds is being processed. Each developed starch-based formulation was constituted by 3 (starch, glycerol, and water) or 4 (in the presence of lipids) compound-types. Therefore, the roughness differences between the films top and bottom surfaces may be related to their random distribution in the polymeric matrix during the solvent evaporation. Moreover, the dispersions viscosity also affects their casting efficiency, thus influencing the films roughness profiles. Therefore, the high viscosity of gelatinized CPS-based dispersions observed at 90 ºC (**Fig. 2**) may also have influenced the solvent evaporation rate, causing a hill-valley-structure, as observed on the CPS-based films top surface. On the other hand, the topographic changes observed between starch/oil and starch/waxes-based films may derive from micelles self-assembling provoked during the homogenization of starch/lipid emulsions that depend on the lipids chemical nature. The oil, constituted by esterified saturated and unsaturated fatty acids (**Fig. S4**) can easily form starch/lipid micelles. In turn, potato peel waxes, formed by phenolic and aliphatic constituents (hydroxyfatty acids, dicarboxylic acids, and very long-chain acids and alcohols), compromise the starch/lipid micelles formation [37].

3.2.3. Wettability

Fig. 4 allows to observe that the water contact angles of neat RPS-based films were similar on both top and bottom surfaces, highlighting their compositional homogeneity and hydrophilicity (40º). In contrary, CPS-based films showed a higher water contact angle on top surface (90º) than on bottom surface (62º), thus being more hydrophobic. This phenomenon may be related with the high viscosity peak of gelatinized CPS-based dispersions (**Fig. 2**) that, during the casting, may hinder the solvent evaporation, originating heterogeneous and rough surfaces, in accordance with **Fig. 3**. The water contact angle increase promoted by oil and waxes on both RPS-based films surfaces was more evident on top surfaces, pointing out the separation between water and lipid phases and/or micelles migration caused by the compounds density differences. This wettability profile was also observed when CPS was used in the films preparation. The oil caused a higher water contact angles variability between the CPS-based films surfaces than waxes. The different chemical composition between oil and waxes, in which the oil triacylglycerides have lower affinity with water molecules than the phenolic and aliphatic constituents present on waxes structure, may explain this behaviour [37].

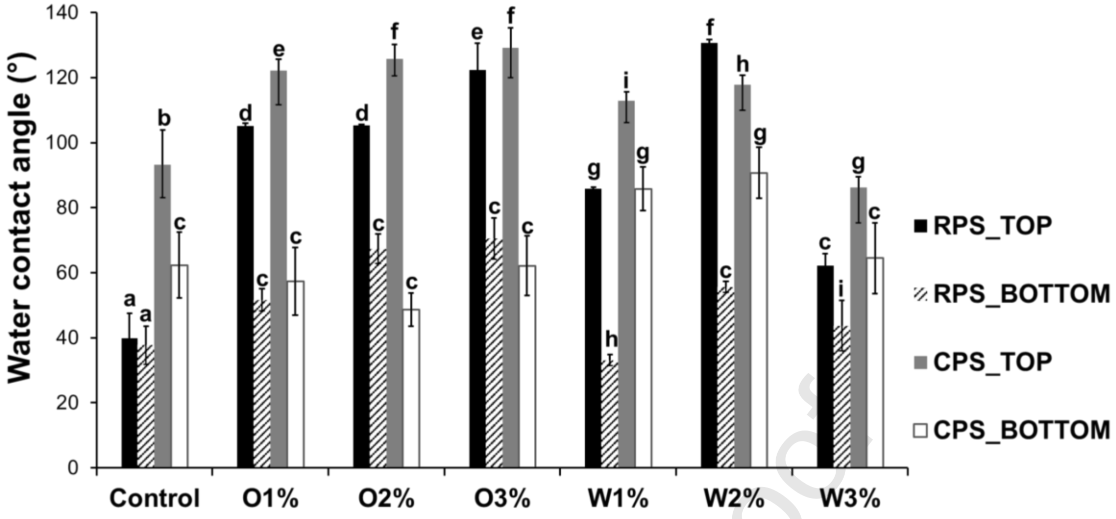

Fig. 4: Water contact angle values of recovered potato starch-rich fraction (**RPS**)- and commercial potato starch (**CPS**)-based films, containing different amounts (0; 1; 2; and, 3% (w/w) of dry starch weight) of oil and waxes. **TOP** and **BOTTOM** are related with the film faces exposed to air and in direct contact with the plexiglass plate during the solvent casting process, respectively.

3.2.4. Mechanical Properties

Fig. 5 demonstrates that, in the absence of lipids, RPS-based films showed higher thickness and elongation at break values, and lower tensile strength and Young's modulus values than CPS-based ones. Therefore, RPS can be used as a raw material that gives rise to starch films with improved plastic properties, which can be explained by the presence of low molecular weight compounds in RPS that can contribute to the films' plasticization. On the other hand, oil and waxes incorporation diminished the thickness of RPS-based films and increased their elongation at break, tensile strength, and Young's modulus values.

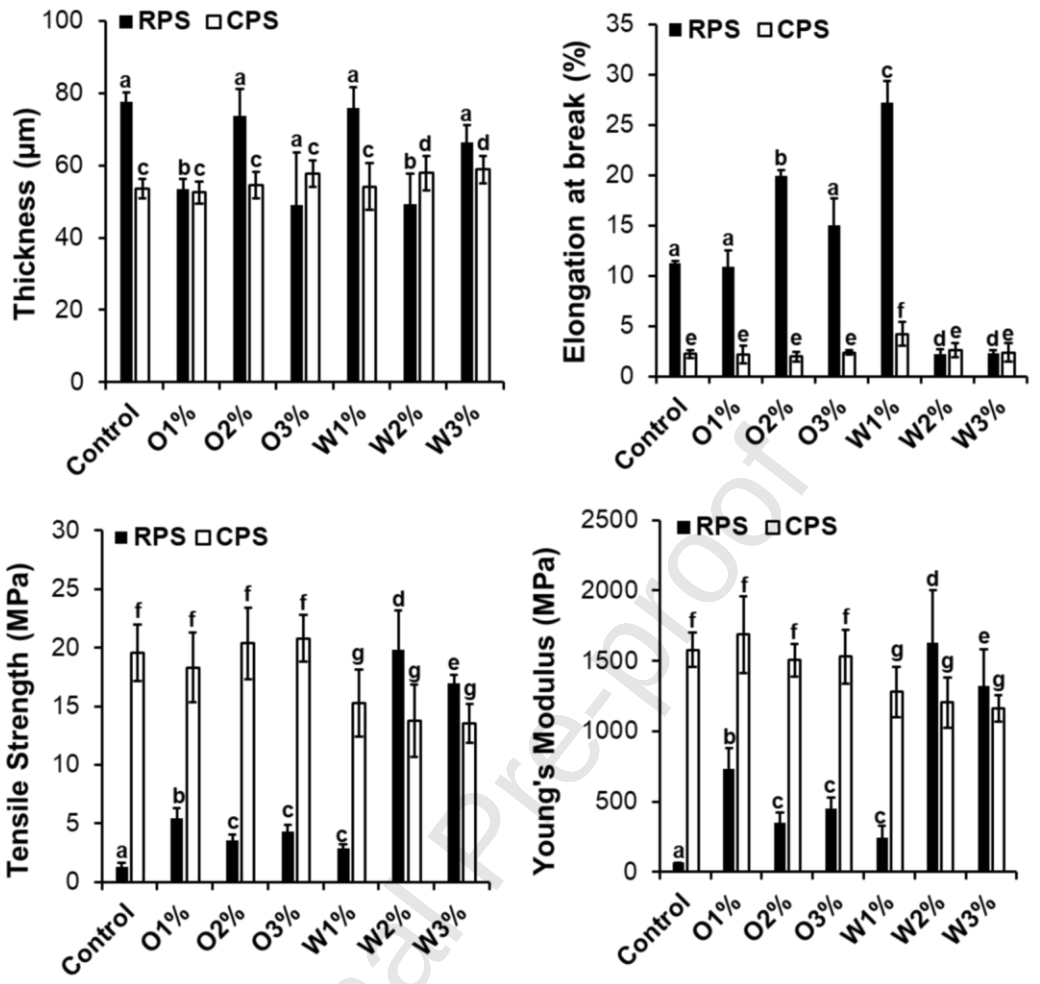

Fig. 5: Thickness, elongation at break, tensile strength, and Young’s modulus of recovered potato starch-rich fraction (**RPS**)- and commercial potato starch (**CPS**)-based films, containing different amounts (0; 1; 2; and, 3% (w/w) of dry starch weight) of oil and waxes.

These mechanical performances can be explained by the starch matrix discontinuity caused by the both lipids studied [38-41], thus decreasing the cohesion forces within the amylose network and impairing mobility to RPS-based films [38, 39]. In CPS-based formulations, only waxes mixture diminished the tensile strength and films Young’s modulus, which may be related to the high viscosity of gelatinized CPS-based formulations that may hinder the lipids dispersion, promoting their self-association within the polymeric structure.

4. Conclusion

Starch recovered from potato washing slurries is suitable for developing biobased plastics, allowing to obtain films with higher plasticity than the films developed with commercially available potato starch. In turn, oil and waxes recovered from potato frying residues and potato peels, respectively, can tailor the surface properties (roughness and wettability) and flexibility of starch-based films. Therefore, potato chips industry byproducts are a rich source of biomolecules of interest for the development of biobased plastics with enhanced physicochemical and mechanical performance, opening an opportunity for their valorisation.

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Figure Captions

Fig. 1: Recovered potato starch-rich fraction (RPS) and commercial potato starch (CPS) granules shape, size, and crystallinity. (A) Scanning electron microscopy micrographs, (B) size distribution, and (C) XRD pattern.

Fig. 2: Pasting profiles of recovered potato starch-rich fraction (RPS) and commercial potato starch (CPS) suspensions (5% w/V) prepared in distilled water.

Fig. 3: 3D AFM topographic images of recovered potato starch-rich fraction (**RPS**) and commercial potato starch (**CPS**)-based films, containing different amounts (0; 1; 2; and, 3% (w/w) of dry starch weight) of oil (**O**) and waxes (**W**). **TOP** and **BOTTOM** are related with the film faces exposed to air and in direct contact with the plexiglass plate during the solvent casting process, respectively.

Fig. 4: Water contact angle values of recovered potato starch-rich fraction (**RPS**)- and commercial potato starch (**CPS**)-based films, containing different amounts (0; 1; 2; and, 3% (w/w) of dry starch weight) of oil and waxes. **TOP** and **BOTTOM** are related with the film faces exposed to air and in direct contact with the plexiglass plate during the solvent casting process, respectively.

Fig. 5: Thickness, elongation at break, tensile strength, and Young’s modulus of recovered potato starch-rich fraction (RPS)- and commercial potato starch (CPS)-based films, containing different amounts (0; 1; 2; and, 3% (w/w) of dry starch weight) of oil and waxes.

Tables

Table 1: Starch content, amylopectin/amylose ratio, and gelatinization properties (onset (To), peak (Tp), and conclusion (Tc) temperatures, and gelatinization enthalpy (ΔH)) of recovered potato starch-rich fraction (RPS) and commercial potato starch (CPS)

	RPS	**CPS**
Starch content (% in dry matter)	72.0 ± 0.8	72.2 ± 1.2
Amylopectin/Amylose ratio	2.3	2.7
Gelatinization properties		
T_o (ºC)	55.1 ± 1.4	58.2 ± 0.8
T_p (ºC)	59.0 ± 0.1	62.0 ± 1.1
T_c (ºC)	71.4 ± 0.1	74.4 ± 0.1
T_c - T_o (ºC)	16.4 ± 1.5	16.2 ± 0.8
ΔH (J/g, db)	11.8 ± 3.6	12.7 ± 1.3

Table 2: Mean values for lightness (*L*),* red-green *(a*),* yellow-blue *(b*),* and total color (ΔE) of recovered potato starch-rich fraction (RPS)- and commercial potato starch (CPS)-based films, containing different amounts (0; 1; 2; and, 3% (w/w) of dry starch weight) of oil (O) or waxes (W). Different letters represent significantly different values ($p < 0.05$)

		L*	***a****	***b****	**ΔE***
RPS	**Control**	91.55 ± 0.36^{a}	0.87 ± 0.04^{b}	-1.85 ± 0.04^{i}	---
	O1%	91.61 ± 0.35^{a}	0.73 ± 0.02^{c}	-1.89 ± 0.02^{i}	0.16
	O2%	90.97 ± 0.37^{a}	0.60 ± 0.06^{d}	-1.08 ± 0.02^{j}	1.00
	O3%	91.14 ± 0.28^{a}	0.59 ± 0.03^{e}	-0.93 ± 0.08^{k}	1.04
	W1%	91.92 ± 0.16^{a}	0.72 ± 0.02^{f,c}	-1.39 ± 0.05^{l}	0.61
	W2%	91.31 ± 0.53^{a}	0.57 ± 0.01^{g,d}	-0.94 ± 0.04^{m,j}	0.99
	W3%	91.26 ± 0.32^{a}	0.45 ± 0.04^{h}	-0.22 ± 0.06^{n}	1.71
CPS	**Control**	92.98 ± 0.15^{o}	0.58 ± 0.02^{r}	-0.21 ± 0.02^{y}	
	O1%	93.02 ± 0.12^{o}	0.29 ± 0.02^{s}	-0.21 ± 0.08^{y}	0.29
	O2%	92.94 ± 0.30^{o}	0.21 ± 0.03^{t}	-0.21 ± 0.07^{y}	0.37
	O3%	93.01 ± 0.43^{o}	0.17 ± 0.05^{u}	-0.20 ± 0.05^{y}	0.41
	W1%	92.88 ± 0.14^{o}	0.40 ± 0.02^{v}	0.21 ± 0.11^{z}	0.47
	W2%	92.34 ± 0.23^{p}	0.22 ± 0.02^{w,t}	0.88 ± 0.13^{A}	1.31
	W3%	92.17 ± 0.14^{q}	0.04 ± 0.05^{x}	1.40 ± 0.34^{B}	1.88

Tailoring the surface properties and flexibility of starch-based films using oil and waxes recovered from potato chips byproducts

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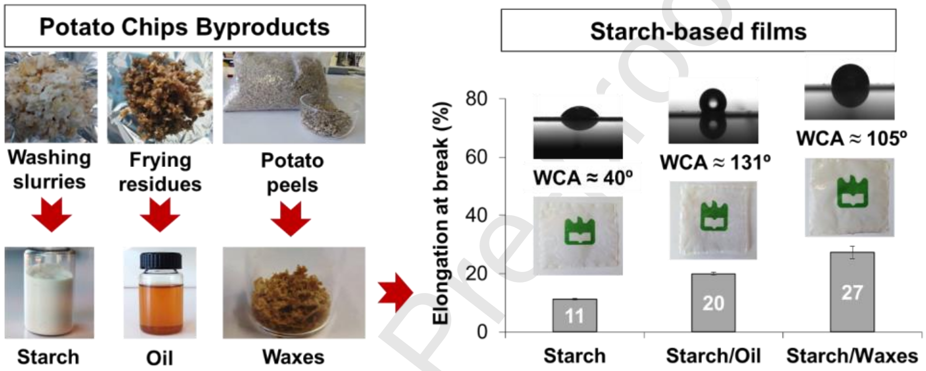

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Highlights

- Non-used byproducts from potato chips industry are valorized.
- Starch and lipidic biomolecules are recovered.
- Transparent starch-based films are produced.
- Oil and waxes improve water tolerance and flexibility of starch films.