

Heat-sealable bioplastic films of blended locust bean and potato byproducts for active packaging of fatty foods: Cheese and oat cookies as case studies

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ABSTRACT

Fatty foods are very susceptible to lipid oxidation caused by oxygen and light. To minimize this issue, active bioplastic materials are suitable for food packaging. In this work, the feasibility of blending locust bean milling derived dust (LBMD) with potato starch to develop heat-sealable bioplastic films for packaging sliced cheese and oat cookies was studied. Blending LBMD with starch allowed to obtain bioplastic films 2-fold more rigid (Young's modulus of *ca.* 48 MPa) and 1.3-fold more resistant to water (water contact angle of *ca.* 87°) than LBMD-based films (Young's modulus of *ca.* 12 MPa and water contact angle of *ca.* 47°), maintaining their UV protective capacity. LBMD/starch-based bioplastic films were used to pack cheese, with no molds or yeasts grown after 21 days, similar to petroleum-based plastic packages used as reference. Despite cheese dehydration (*ca.* 10% weight loss), its texture did not significantly change after the storage period. An increase (*ca.* 43%) in volatile compounds derived from cheese oxidation was observed after 7 days of storage. When used to pack oat cookies, LBMD/starch-based bioplastic films played an active role in decreasing oxidation-derived volatile compounds after 21 days of storage, maintaining their textural properties, when compared to oat cookies packaged with petroleum-based plastic. Therefore, blending LBMD with starch revealed to be a proper strategy to develop heat-sealable and active bioplastic films with water absorption, protection against UV radiation, volatile compounds scavenging capacity, and antioxidant activity suitable to preserve fatty foodstuffs while providing them new characteristics.

1. Introduction

Packaging represents an important protection for foodstuffs, allowing to preserve the quality and extend their shelf-life. Fatty foods require protection from light and oxygen to prevent lipid oxidation and consequent deterioration of food quality and safety (Mestdagh et al., 2005). Therefore, the choice of packaging materials must consider the mechanical resistance and barrier to water vapor and oxygen (Pires et al., 2021), as well as the storage temperature and humidity conditions (Chen et al., 2019). These protections are often achieved through non-biodegradable petroleum-based plastic packaging. Aiming to decrease the environmental footprint of single-use food packaging while minimizing food loss, active biobased materials with enhanced mechanical and barrier properties and the ability of actively prevent lipid

oxidation have been explored. Natural biopolymers, such as proteins and polysaccharides, have been considered for this purpose (Castro et al., 2019; Lopes et al., 2021, 2023; Martínez et al., 2013; Siripatrawan & Vitchayakitti, 2016; Yadav et al., 2020). In addition, essential oils or extracts of phenolic compounds obtained from plants confer antioxidant properties (Lee et al., 2023; Marturano et al., 2019). Gelatin- and whey protein-based bioplastic films combined with antioxidant and antimicrobial plant extracts have shown the suitability to be used as coatings and packages for cheese (Jridi et al., 2020; Ramos et al., 2012), and salmon (Castro et al., 2019). These bioplastics prevented microbial growth for at least 7 days while increasing the firmness of cheese texture (Jridi et al., 2020; Ramos et al., 2012) and delaying salmon lipid oxidation for up to 14 days (Castro et al., 2019). The addition of essential oils and plant extracts to chitosan-based bioplastic films showed the

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ability to protect pork (Bonilla et al., 2014) and cheese (de Elguea-Culebras et al., 2019) against lipid oxidation and microbial growth. Nevertheless, due to the lack of heat-sealing ability of most protein- and polysaccharide-based materials, many of these studies only pack the foods by wrapping, which allow the oxygen permeation through the package (Jridi et al., 2020; Ramos et al., 2012). To overcome this drawback, the blend with thermoplastic biomolecules have been considered.

Starch is a biomolecule constituted of amylose and amylopectin, two polysaccharides that gelatinizes in the presence of plasticizers and temperature, allowing to develop thermoplastic biobased materials (Zhang et al., 2014). Due to this feature, starch-based materials can be processed using the most conventional polymer processing techniques and heat-sealed (Jiang et al., 2020), being a fundamental condition for the application of films as food packaging. When combined with gelatin (Izzi et al., 2023), chitosan (Alves et al., 2022; Bof et al., 2021) and pea protein (Huntrakul et al., 2020), starch allowed to obtain bioplastic films with heat-sealing properties.

To minimize the use of edible resources in the development of biodegradable food packaging, non-valued agrifood byproducts rich in proteins and/or polysaccharides have been considered (Araújo et al., 2018; Felix et al., 2017; Lopes et al., 2021; Omrani-Fard et al., 2020; Pérez-Bassart et al., 2023). Starch, as well as proteins, other polysaccharides, and phenolic compounds can be easily found in non-edible and often wasted agrifood byproducts, such as the powder resulting from milling and sieving locust bean seeds (Lopes et al., 2023) and potato washing slurries (Gonçalves et al., 2020). Therefore, its valorization through the development of heat-sealable and active bioplastic materials, taking advantage of the antioxidant activity resulting from the presence of phenolic compounds, is an opportunity for fatty foods packaging.

In this work, it is hypothesized that blending locust bean milling dust (LBMD) with starch recovered from potato washing slurries allows to develop a heat-sealable bioplastic film suitable for fatty food packaging. To validate this hypothesis, the physicochemical, mechanical, and barrier properties of LBMD/starch-based bioplastic films were evaluated. Then, LBMD/starch-based packages of cheese and oat cookies, two fatty food products with different physical state and water content that require different storage conditions, were developed, and their chromatic properties, texture, odor, flavor, and oxidation-derived volatile compounds were analyzed over 21 days.

2. Materials and methods

2.1. Samples and reagents

Locust bean milling dust (LBMD) (56% protein, 28% polysaccharides, 6% lipids, and phenolic compounds (Lopes et al., 2023), thermal degradation at ca. 205 °C) was provided by Industrial Farese, Lda. company (Faro, Portugal). Potato starch (70% amylopectin (350 kDa) and 30% amylose (50 kDa) (Gonçalves et al., 2020), thermal degradation at ca. 275 °C) was obtained from potato washing slurries and was provided by A Saloinha, Lda. company (Mafra, Portugal). Glycerol (99.5%) was supplied by Scharlab S. L. (Barcelona, Spain). Sodium azide was distributed from Panreac Quimica SAU (Barcelona, Spain). Anhydrous calcium chloride (97%) was provided from Sigma-Aldrich (St. Louis, MO, USA). Dichloran-rose bengal chloramphenicol (DRBC) agar Base (oxidoid CM0727) was order from Oxoid (Hampshire, England). Heptanoic acid ethyl ester was obtained from Fluka (Buchs, Switzerland), and methanol from Supelco (Bellefonte, Pennsylvania). All the used reagents were of analytical grade.

2.2. Preparation of LBMD/starch-based bioplastic films

The bioplastic films were prepared according to the method described (Lopes et al., 2021). The blend of LBMD (5% w/V) and starch

(2% or 4% w/V) were dispersed in distilled water and gelatinized (95 °C) in the presence of glycerol (30% w/w related to biopolymeric portion dry weight), with continuous stirring (300 rpm), during 30 min. The solutions were poured onto plates, dried in an oven (Venticell Eco line, MMM Group, Germany) at 25 °C with circulating air, and the films were removed from the plates. During the drying of the films, two different surfaces were obtained, the top surface exposed to air during drying and the bottom surface in contact with the acrylic plate. Neat LBMD- and starch-based bioplastic films were prepared in the same way and used as controls. After drying, the samples were stored at 53% relative humidity for at least 5 days for further characterization.

2.3. Characterization of LBMD/starch-based bioplastic films

2.3.1. Chromatic, water tolerance, morphological and mechanical properties

Chromatic properties of the bioplastic films were determined by CIELab chromatic parameters, L^* , a^* , and b^* using a Minolta CM-508d colorimeter (Minolta, Kyoto, Japan) (Lopes et al., 2021). Water wettability was evaluated by measuring the water contact angle (WCA) between 3 μ L of ultrapure water drop and bioplastic surface using a Attension Theta optical tensiometers (Biolin Scientific, Västra Frölunda, Sweden). Bioplastic films water solubility was determined by weight loss measurements over 7 days, at room temperature with 80 rpm orbital shaking (ELMI DOS-20 L, Riga, Latvian). Tensile strength, Young's modulus, and elongation at break of bioplastic films were determined by performing tensile tests in a TA. XTplusC texture analyzer (Stable Micro Systems, Maia, Portugal) and calculated according to the reported equations (Gonçalves et al., 2020). The surface morphology and cross-section of the bioplastic films was observed by high-resolution scanning electron microscopy (SEM) using a Hitachi SU-70 microscope (Hitachi High-Tech Corporation, Tokyo, Japan) operated at 4 kV and 8.7 mm of working distance. The sample was coated with carbon.

2.3.2. Gas and light barrier and migration tests

Water vapor transmission rate (WVTR in $\text{g H}_2\text{O}/\text{m}^2\cdot\text{day}$) was gravimetrically determined using cylindrical plexiglass cells covered with a circular film specimen, following the ASTM-E96 (Lopes et al., 2021). Permeation cells containing anhydrous calcium chloride inside to promote water vapor diffusion through the bioplastic film were used, under two different conditions: (1) 23 °C and 50% relative humidity and (2) 4 °C and 80% relative humidity. Each measurement was performed on at least 3 independent replicates. The temperature and relative humidity conditions were selected to mimic the storage conditions of the packaged products (cheese and oat cookies).

The oxygen transmission rate (OTR in $\text{mL O}_2/\text{m}^2\cdot\text{day}$) and the carbon dioxide transmission rate (CO_2TR in $\text{mL CO}_2/\text{m}^2\cdot\text{day}$) were determined according to the previously described procedure (Larsen et al., 2000; Larsen & Liland, 2013). The permeability cells consisted of a cylinder that was covered with a circular bioplastic film specimen and were flushed with the gas mixture 0.2% O_2 and 20% CO_2 in N_2 (Linde ÅS, Oslo, Norway). Each cell was placed in a test chamber containing air of atmospheric composition, with controlled temperature and relative humidity, using two different conditions: 23 °C and 50% relative humidity and 4 °C and 80% relative humidity, with 0% internal humidity. The O_2 and CO_2 values were periodically measured using a PBI Dan-sensor Checkmate II headspace analyzer (Ametek Mocon, Minneapolis, USA). The measurements were performed in triplicate.

UV-protective capacity was evaluated at absorbance wavelength ranging from 190 nm to 900 nm. using a GBC Cintra 303 UV-visible spectrophotometer (GBC Scientific Equipment Ltd, Dandenong, Australia).

Migration tests using simulant recommended for foods with high content in fat, namely 95% ethanol and isooctane (Bolognesi et al., 2016), were carried out as defined for plastic materials intended to be used in contact with food (European Commission, 2011).

LBMD/starch-based bioplastic film squares (11 cm²) were immersed into 95% ethanol and kept at 40 °C for 10 days. In the case of isoctane, the analysis was carried out at 20 °C for 2 days. Each analysis was performed in triplicate.

2.4. LBMD/starch bioplastic films as food packaging for cheese and oat cookies

2.4.1. Packaging and storage of cheese and oat cookies

Thirty individual LBMD/starch-based bioplastic film bags containing 6 slices (≈100 g) of pre-sliced Norvegia® original cheese were made using a vacuum-sealing packaging machine, a Henkelman Boxer 42XL (CK 's-Hertogenbosch, Netherland), and sealing for 1.5 s. The samples were stored at 4 °C with 80% relative humidity (RH). Polyamide/polyethylene (PA/PE 20/70) (20 μm PA/70 μm PE, Allfo Vakuumverpackungen, Waltenhofen, Germany) plastic, commonly used in food packaging, was used as a control. The cheese samples were analyzed after 7, 14, and 21 days of storage.

To evaluate the potential for the LBMD/starch-based material for a product with high fatty content but low water content as well as at different storage temperature and humidity condition, oat cookies were packaged. Fifteen individual packages with 2 BIXIT® commercially available oat cookies were sealed in LBMD/starch-based packaging using a Packer sealing machine (Packer Poly Sealer, Norfolk, England) for 1.5 s. PA/PE-based packaging was also prepared, under the same conditions. Packages were stored at room temperature (20 ± 5 °C) and relative humidity (40% ± 5%). Samples were analyzed after 7 and 21 days of storage.

2.4.2. Food quality analyses of cheese and oat cookies

2.4.2.1. General physicochemical analysis. The color, given as *L** (lightness), *a** (red-green), and *b** (yellow-blue) parameters was measured by using a Minolta Chromameter CR-400 (Minolta Konica Sensing Inc., Osaka, Japan) with an 8 mm viewing port, 2° viewer angle, and illuminant D65. The color was measured in five spots of each sample and for all replicates (3). The results were given as color variation (ΔE) (Lopes et al., 2021).

Cheese weight loss (given as % weight loss) was gravimetrically monitored during storage, using a Mettler Toledo NewClassic MS precision scale (Mettler Toledo, Greifensee, Switzerland).

Cheese pH was determined on day 0 and after 21 days of storage, in triplicate, by diluting the cheese in distilled water using a Knick pH meter (Knick GmbH & Co, Berlin, Germany).

2.4.2.2. Texture analysis. Texture changes through the calculation of puncture work were evaluated using a TA. XTplus100C texture analyzer equipment (Stable Micro Systems, Surrey, United Kingdom) with a cylindrical probe of 6 mm diameter fitted to the cell (crosshead speed 1 mm s⁻¹) (Benedito et al., 2006). Ten measurements in 3 replicates were performed for each sampling time and the data was analyzed using the software Exponent version: 6.1.16.0. Results were given as puncture work in N.mm.

2.4.2.3. Volatile compounds analysis. Formation of oxidation products developed during storage was analyzed by headspace/gas chromatography-mass spectrometry (HS/GC-MS). The volatile compounds were extracted at 70 °C, for 20 min, in the presence of the internal standard heptanoic acid ethyl ester. Volatile compounds were trapped through an adsorbent tube packed with Tenax GR 60/80. Volatiles were desorbed in a Markes Unity/Ultra TD automatic desorber (Markes International ltd, Lantisant, England) and then separated on an Agilent 6890 GC (Agilent, Palo Alto, CA, USA) with an Agilent 5973 mass selective detector, and a DB-WAXetr column, using a described temperature program (Sarfranz et al., 2021). The peaks obtained were

integrated, identified, and the results were expressed as μg of ethyl heptanoate equivalents (eq.)/g of sample.

2.4.2.4. Microbial analysis. Microbial analyses of cheese, identifying the presence of molds and yeasts, were analyzed at the time of packaging and at each sampling time (7, 14, and 21 days). For this, 1 slice of cheese was weighed, diluted 1:10 with peptone water, and macerated 1 min (NMKL No 91, 2010). The samples were spread on plates with dichloran-rose bengal chloramphenicol (DRBC) agar base (oxid CM0727) medium and incubated at 25 °C, for 5–7 days (NMKL No 98, 2005). Analyses were performed in triplicate.

2.4.2.5. Odor and flavor evaluation. Evaluation of odor and flavor was performed at each sampling time by lab panel consisting of 5 semi-trained assessors. Prior to the evaluation the assessors were trained using the actual method and agreement in freshness (highest intensity of fresh odor and flavor) was discussed. The evaluation included the slices that had been in contact with the packaging material, thus only the top and bottom slices in the packages. The samples for evaluation were cut, prepared, and stored in room temperature 30 min prior to the evaluation. Samples were equal-sized and coded with a three-digit number following a randomized block design. The assessors evaluated the odor first and then the flavor. Four replicates of each sample were evaluated at each sampling time. The samples were ranked on a scale of 1–5, with score 5 indicating highest intensity of fresh flavor and odor and highly acceptable/good quality, a score of 3 indicates perceived alteration of the sample but still acceptable, while score 1 indicates a highly perceived and not acceptable freshness (Fig. S1A for cheese and Fig. S1B for oat cookies). At each sampling time, the sample in the original package was evaluated, in addition to the sample packaged in LBMD/starch-based bioplastic film and PA/PE-based plastic. The results were reported as the average of scores. Cheese attributes, such as pungency, acidity, hardness, and rancidity, were also qualitatively evaluated by the panelists. For oat cookies, sweetness, hardness, and rancidity were evaluated.

2.5. Statistical analysis

Statistical analyses were performed using analysis of variance (ANOVA) and Tukey's HSD post hoc test, at a 95% of significance level, using IBM SPSS Statistics Software (IBM Corp. Released 2019. IBM SPSS Statistics for Windows, Version 26.0. Armonk, NY, EUA).

3. Results and discussion

LBMD/starch-based bioplastic films were developed to take advantage of the thermoplastic behavior of starch and the flexibility of LBMD bioplastic films, and their chromatic properties, water solubility, wettability, mechanical, and gas barrier properties were characterized. The developed LBMD/starch-based bioplastic films, due to their sealable characteristics and improved physicochemical, mechanical and barrier properties, compared to LBMD- or starch-based bioplastic films, were applied in food packaging, using cheese and oat cookies as exemplary foods. These foods were selected as they are rich in fat but different in water content and stored in different humidity and temperature conditions.

3.1. Characterization of LBMD/starch-based bioplastic films

3.1.1. Chromatic, water tolerance, morphological, and mechanical properties

When combined with 4% starch, the 5% LBMD-based suspension showed a high viscosity, thus compromising the bioplastic film production, as compared to the solution containing 2% starch. Therefore, all the bioplastic films characterized in this work were constituted by 5%




LBMD, 2% starch, water, and glycerol. Blending starch with LBMD had lower L^* (83.68) and a^* (−4.71) values but higher b^* (16.84) value (Table 1) than LBMD-based films. Thus, the incorporation of starch intensified the yellowish coloration of 5% LBMD-based bioplastic films, although without compromising their transparency. Nevertheless, the total color variation (ΔE) of LBMD/starch-based films changed only 2.48, when compared to neat LBMD-based bioplastic films, meaning that this variation was not detectable by the average human eye. The yellowish coloration provided UV radiation protection to the bioplastics, as observed by absorbance values (190 nm–290 nm) ranging from 1.3 to 9, corroborating previously reported for soy protein-based films (Li et al., 2021).

The thickness, tensile strength, Young’s modulus, and elongation at break of the obtained LBMD/starch-based bioplastic films are presented in Table 2.

The bioplastics films prepared from 5% LBMD and 2% starch were characterized for comparison purposes. Adding 2% of starch increased the 5% LBMD bioplastic film’s thickness and Young’s modulus from 75 μm to 139 μm and 24 MPa–48 MPa, while decreasing the elongation at break from 87% to 64% of LBMD-based bioplastic films without changing their tensile strength (5 MPa). Therefore, starch conferred rigidity to the LBMD-based bioplastic film. When compared to the materials prepared using 2% starch, LBMD/starch-based bioplastic films were more rigid (Young’s modulus of 12 MPa) and stretchable

Table 1

CIELab values of cheese and oat cookies packaged with LBMD/Starch-based bioplastic films and non-biodegradable PA/PE-based plastic, on different sampling days. CIELab values of LBMD/starch-, starch- and LBMD-based bioplastic films. Different letters represent values significantly different within a storage time and product (cheese and oat cookies), as well as in bioplastic films ($p < 0.05$).

Cheese	Time (days)	L^*	a^*	b^*	ΔE
	0	81.35 \pm 0.59 ^a	−2.80 \pm 0.11 ^a	30.87 \pm 0.37 ^a	–
LBMD/ Starch	7	74.47 \pm 2.71 ^a	−2.79 \pm 0.05 ^a	35.12 \pm 1.17 ^a	8.12 \pm 2.87 ^c
PA/PE		80.42 \pm 0.23 ^{a,b}	−2.72 \pm 0.02 ^a	31.76 \pm 0.33 ^{a,b,c}	1.29 \pm 0.38 ^{a,b}
LBMD/ Starch	14	73.12 \pm 2.80 ^c	−2.57 \pm 0.16 ^{a,b}	36.15 \pm 0.96 ^d	9.82 \pm 2.80 ^c
PA/PE		80.93 \pm 1.13 ^{a,b}	−2.39 \pm 0.07 ^b	32.25 \pm 0.32 ^{b,c}	1.73 \pm 0.66 ^{a,b}
LBMD/ Starch	21	78.12 \pm 0.53 ^b	−3.30 \pm 0.28 ^c	32.84 \pm 0.51 ^c	3.86 \pm 0.36 ^b
PA/PE		82.15 \pm 0.27 ^a	−2.67 \pm 0.14 ^{a,b}	31.27 \pm 0.18 ^{a,b}	0.94 \pm 0.21 ^a
Cookies	Time (days)	L^*	a^*	b^*	ΔE
	0	44.63 \pm 1.13 ^a	15.10 \pm 0.56 ^a	30.91 \pm 0.37 ^a	–
LBMD/ Starch	7	46.96 \pm 1.44 ^a	14.34 \pm 0.89 ^a	32.82 \pm 0.63 ^{b,c}	3.30 \pm 1.30 ^{b,c}
PA/PE		44.62 \pm 1.44 ^a	15.03 \pm 0.96 ^a	31.45 \pm 0.54 ^a	1.52 \pm 0.86 ^{a,b}
LBMD/ Starch	21	47.03 \pm 1.44 ^a	14.61 \pm 0.90 ^a	33.42 \pm 1.02 ^c	3.62 \pm 1.70 ^c
PA/PE		45.13 \pm 0.73 ^a	14.92 \pm 0.49 ^a	31.53 \pm 0.84 ^{a,b}	1.10 \pm 0.89 ^a
	Digital image	L^*	a^*	b^*	ΔE
LBMD/ Starch		83.68 \pm 0.24 ^a	−4.71 \pm 0.02 ^a	16.84 \pm 0.09 ^a	2.48 \pm 0.24 ^a
Starch		91.48 \pm 0.33 ^c	0.87 \pm 0.07 ^c	−1.87 \pm 0.06 ^c	19.08 \pm 0.09 ^b
LBMD ^A		86.11 \pm 0.49 ^b	−4.43 \pm 0.02 ^b	15.66 \pm 1.03 ^b	–

(elongation at break of 3%), although a decrease in the tensile strength (20 MPa) was observed. The increase in rigidity and stretchability was also observed by other authors in films obtained by blending starch and gelatine (Acosta et al., 2015). The increased rigidity of LBMD/starch-based bioplastic films can be explained by the high miscibility and hydrogen bonding interaction between the protein and polysaccharides (Zárate-Ramírez et al., 2014; Zhou et al., 2008), also described for starch and LBMD (Lopes et al., 2021, 2023).

Blending starch with LBMD increased the hydrophobicity of the bioplastic films, reaching water contact angles (WCA) values close to the one recognized as hydrophobic (90°) (Law, 2014) and higher (87°) than those observed in samples exclusively prepared with LBMD (66°) or starch (47°) (Table 2). For the bottom surface, the blending did not have the same effect, as the WCA value (48°) was not significantly different to the ones obtained for 5% LBMD-based bioplastic films (50°) but increased in relation to the 2% starch-based bioplastic films (35°). Differences in top and bottom surfaces wettability may result from the higher heterogeneity of the films top surface, as observed in the scanning electron microscopy micrographs (Fig. 1). These variances in films roughness may be related to the top surface being exposed to air, allowing the solvent evaporation over the time. This leads to the random distribution of film constituents where the more soluble ones are separated from those that tend to precipitate, resulting in higher heterogeneity (Gonçalves et al., 2020). As this phenomenon does not occur in the bottom surface, in contact with the plate, a higher surface uniformity was observed. The increase in WCA may be due to the enrichment of the hydrogen bonding between proteins and polysaccharides, thus decreasing the number of free hydroxyl groups available to interact with water molecules (Zaleska et al., 2000). The increase in WCA was also observed in films based on starch and whey protein, where the authors obtained a WCA of 86° (Basiak et al., 2017). On the other hand, evaporation of water from the top surface can form a cohesive structure that retains water and hydrophilic compounds at the bottom surface, leading to different properties on these two bioplastic film surfaces (Gonçalves et al., 2020). Regarding bioplastic films water sensitivity, adding starch to LBMD decreased the water solubility of bioplastic films from 46% to 28%, when compared to 5% LBMD-based bioplastic films, showing the interaction of LBMD components with starch, which corroborates with the Young’s modulus and WCA. This interaction can also be observed in scanning electron (SEM) micrographs (Fig. 1) in which the decrease in free spaces between the molecules of LBMD/starch-based bioplastic films is noted.

3.1.2. Gas barrier and migration analysis

The gas barrier properties, namely water vapor, oxygen, and carbon dioxide, of the obtained LBMD/starch-based bioplastic films are presented in Table 3.

For the 2 tested temperature and relative humidity conditions (23 °C, 50% RH and 4 °C, 80% RH), the incorporation of starch decreased the water vapor transmission rate (WVTR) values (60 and 284 g H₂O/m². day) of LBMD (221 and 338 g H₂O/m². day), although showing higher WVTR at 23 °C, 50% RH than the 2% starch-based bioplastic films (48 g H₂O/m². day). The same effect was verified for O₂ and CO₂ at 23 °C and 50% RH. However, it was not possible to evaluate the oxygen transmission rate (OTR) and carbon dioxide transmission rate (CO₂TR) at 4 °C for films prepared exclusively with starch or LBMD, since the samples showed cracks on their surface during the analysis, due to their lack of mechanical under these conditions, contrary to LBMD/starch-based bioplastic films. The improvement in gas barrier properties is in line with the materials stiffness increase, indicating a decreased free hydrodynamic volume between LBMD, starch, and the plasticizers (water and glycerol) (Coughlan et al., 2004) that hinders the gas molecules passage across the bioplastic film. Nevertheless, when exposed to lower temperature (4 °C) and 80% RH storage conditions, the gas barrier properties of bioplastics were compromised as a result of the samples hydration. Water molecules can act as plasticizers, leading to an opening

Table 2

Thickness, tensile strength, Young's modulus, and elongation at break, water contact angle of top and bottom surfaces, and water weight loss properties of LBMD/starch-, starch-, and LBMD-based bioplastic films. Different letters represent values that are significantly different ($p < 0.05$).

	Thickness (μm)	Tensile Strength (MPa)	Young's Modulus (MPa)	Elongation at break (%)	Water contact angle		Weight loss (%)
					Top	Bottom	
LBMD/Starch	139.4 \pm 6.0 ^a	4.3 \pm 0.9 ^a	48.5 \pm 12.9 ^a	64.5 \pm 10.0 ^a	87.3 \pm 3.2 ^a	47.6 \pm 3.0 ^a	27.8 \pm 0.5 ^a
Starch	39.6 \pm 6.2 ^c	19.5 \pm 5.3 ^b	12.1 \pm 1.2 ^c	2.6 \pm 0.6 ^c	46.6 \pm 4.0 ^c	34.9 \pm 3.5 ^b	14.7 \pm 0.6 ^c
LBMD ^A	74.7 \pm 3.2 ^b	5.0 \pm 0.5 ^a	24.1 \pm 2.4 ^b	87.2 \pm 3.7 ^b	65.9 \pm 1.5 ^b	49.8 \pm 1.4 ^a	46.0 \pm 0.5 ^b

^A Data from (Lopes et al., 2023).

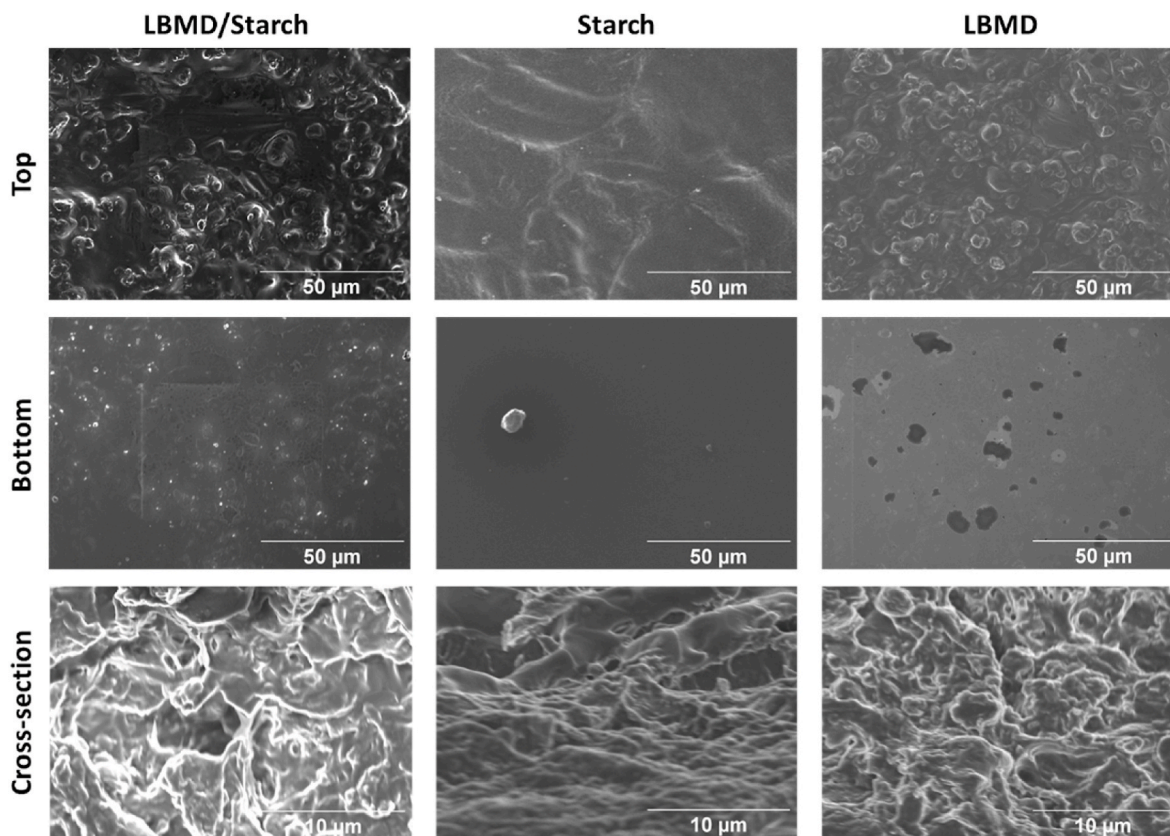


Fig. 1. SEM micrographs of LBMD/starch-, starch-, and LBMD-based bioplastic films for the top, bottom, and cross-section surfaces.

Table 3

Water vapor (WVTR), oxygen (O₂TR), and carbon dioxide (CO₂TR) transmission rate of LBMD/starch-, starch-, and LBMD-based bioplastic films exposed to different temperature and relative humidity conditions (23 °C, 50% RH and 4 °C, 80% RH; 0% internal humidity). Different letters represent values that are significantly different ($p < 0.05$).

Sample	WVTR (g H ₂ O/m ² . day)		OTR (mL O ₂ /m ² . day)		CO ₂ TR (mL CO ₂ /m ² . day)	
	23 °C	4 °C	23 °C	4 °C	23 °C	4 °C
	50% RH	80% RH	50% RH	80% RH	50% RH	80% RH
LBMD/Starch	60 \pm 5 ^a	284 \pm 14 ^a	17 \pm 1 ^a	1931 \pm 33	188 \pm 54 ^a	52,547 \pm 679
Starch	48 \pm 5 ^b	338 \pm 62 ^b	10 \pm 4 ^b	n.m.	135 \pm 19 ^a	n.m.
LBMD	221 \pm 24 ^c	338 \pm 15 ^c	33 \pm 5 ^c	n.m.	556 \pm 73 ^b	n.m.

n.m. – non-measured due to sample cracks.

of the original compact structure of the material, thus facilitating the diffusion of water vapor, O₂, and CO₂ through the polymeric matrix, as observed for polylactic acid-based bioplastics (Holm et al., 2006; Petersen et al., 2011).

When exposed to simulants recommended for foods with high fat content, namely 95% ethanol and isooctane, LBMD/starch-based bioplastic films became opaque. Moreover, for 95% ethanol, the intensity of their neat yellowish coloration became more lighter, changing to a beige color (Fig. 2A), which was not observed for the bioplastic films immersed in isooctane (Fig. 2B). These observations may be explained by the loss of glycerol when ethanol was used as simulant, which contrasted with lower amount of compounds that migrated to the isooctane phase.

The residue of 95% ethanol obtained after the solvent evaporation showed a very viscous solution, characteristic of the glycerol presence, making difficult it complete drying, and, therefore, it was not possible to determine the overall migration of LBMD/starch-based bioplastic films in 95% ethanol. Moreover, the estimation of the films overall migration in isooctane was not possible due to the obtention of a weight lower than the limit of quantification.

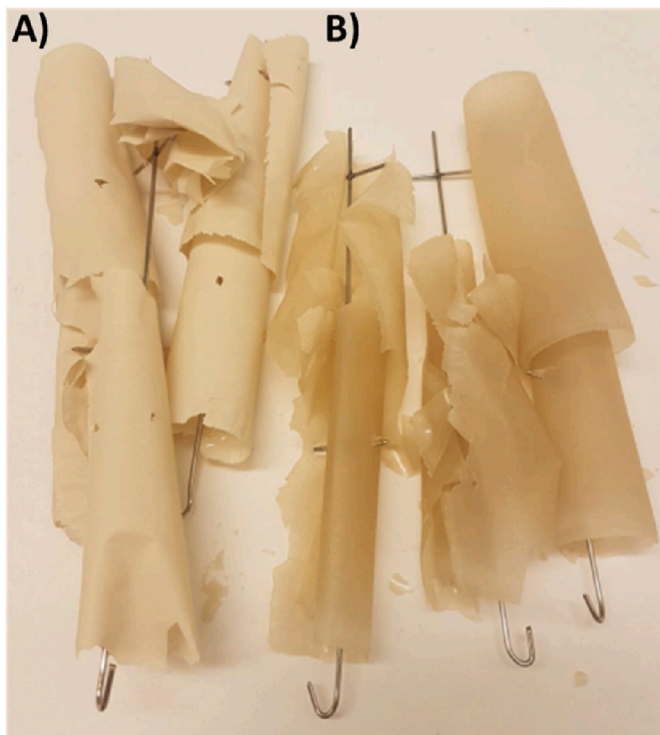


Fig. 2. LBMD/starch-based bioplastic films after immersion in food simulants: (A) 95% ethanol and (B) isooctane.

3.2. Packaging of sliced cheese

Cheese packaged with LBMD/starch-based bioplastic films was evaluated after 7, 14, and 21 days after packaging. Digital images of packages are shown in Fig. 3A.

During the storage period, the cheese packages became wrinkled which means that, although the low water wettability (Table 2), LBMD/starch-based materials still did not show water tolerance to high

moisture conditions as the ones used in the refrigerated storage of sliced cheese.

3.2.1. General physicochemical analysis

The color of cheese packaged with LBMD/starch-based bioplastic films and PA/PE-based plastic was evaluated over time and the results are shown in Table 1. Despite the migration of glycerol in 95% ethanol solution (Fig. 2), during the storage time it was not possible to observe discernible absorption of glycerol by the human eye when opening packages with cheese.

Over time, the lightness (L^*) of cheese packaged in bioplastic films decreased, while the yellow-blue value (b^*) increased, without changing the red-green value (a^*), when compared to the initial cheese (0 days). Therefore, the yellowish coloration of cheese became more intense, contrary to what was observed for cheese packaged in the PA/PE-based plastic, where the cheese neat coloration was maintained. The color change of cheese packaged in LBMD/starch-based bioplastic films may be related to the bioplastics water absorption, but also to the water

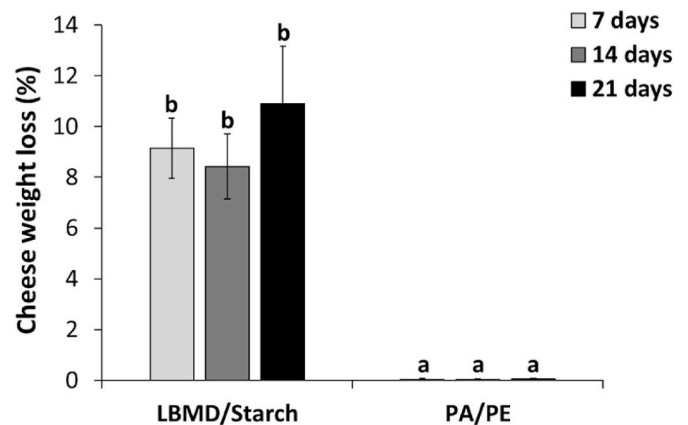


Fig. 4. Cheese percentage weight loss after 7, 14, and 21 days packaged with LBMD/starch-based bioplastic films and non-biodegradable PA/PE-based plastic. Different letters represent values that are significantly different ($p < 0.05$).

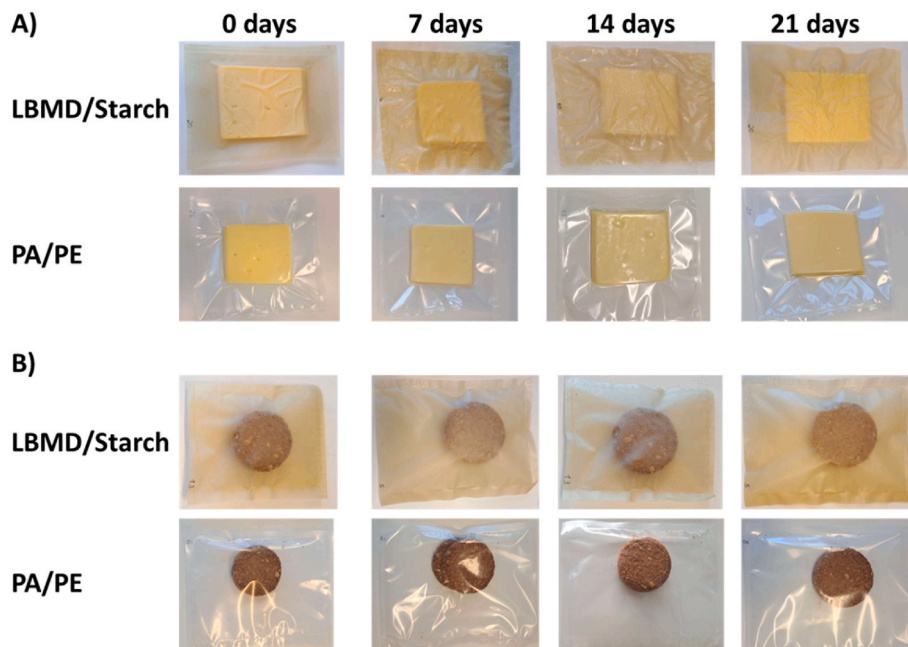


Fig. 3. Digital images of cheese (A) and oat cookies (B) packaged with LBMD/starch-based bioplastic films and non-biodegradable PA/PE-based plastic after 0, 7, 14, and 21 days.

vapor permeation through the bioplastics, leading to a partial dehydration of the cheese (Fig. 4) and intensification of its yellow color (Holm et al., 2006; Lopes et al., 2021). The weight of bioplastics increased 6% in 7 days while the cheese dehydration increased 9%, supporting the water permeation through the bioplastics. After 7 days, the cheese lost 9% of its weight (Fig. 4), without changing until the 21 days, showing a tendency towards stabilization between the cheese and the LBMD/starch-based bioplastic films. Increased yellow color and cheese dehydration was also observed in cheese packaged with gelatin-based bioplastic films (Jridi et al., 2020). Cheese dehydration may also be associated with the barrier properties of the bioplastic films (Table 3). The dehydration effect was not observed for cheese packaged with PA/PE-based plastics, as they present better water vapor barrier properties ($2.1 \pm 0.1 \text{ g H}_2\text{O/m}^2 \cdot \text{day}$ (Vytejčková et al., 2017)).

3.2.2. Texture analysis

Puncture work or the work performed by the probe to perforate the cheese is represented in Fig. 5. This texture parameter allows evaluating the hardness of cheese during storage time. Cheese hardness tends to increase over time when cheese was packaged in LBMD/starch-based bioplastic films, contrary to the PA/PE-based packaging, however the increase is not statistically significant. This texture change can be related to cheese dehydration phenomenon, as observed in cheese packaged with chitosan-based bioplastic films (Sabbah et al., 2019).

3.2.3. Microbial analysis

The microbiological analysis carried out on the cheese showed that

at the initial time, as well as after 7, 14, and 21 days of storage, there was no appearance of molds or yeasts in any of the cheese packages.

3.2.4. Odor and flavor evaluation

The influence of LBMD/starch-based bioplastic films on the odor and flavor of cheese during storage time was evaluated by a semi-trained lab panel (Fig. 6). Considering the odor, changes were perceived in both packages, although acceptable for 14 days (evaluation score higher than 3). This difference was more pronounced in the sample packaged in bioplastic, which can be related with the loss of water vapor and O_2 of LBMD/starch-based bioplastic films at the refrigerated conditions (Table 3), bringing up oxidation reactions and formation of perceived volatile compounds. After 7 days, the flavor of the cheese packaged with LBMD/starch-based bioplastic film was statistically similar to the original package, contrary to the cheese packaged in PA/PE-based plastic. After 14 days the changes although acceptable were more noticed in LBMD/starch-based bioplastic films than in PA/PE-based plastic. Cheese attributes, such as pungency, acidity, hardness, and rancidity, were also qualitatively evaluated by the panelists. None of the samples showed changes in pungency, during the whole storage time, when compared to the neat cheese (day 0).

The perception of cheese acidity decreased along the time, possibly related to the loss of cheese smell, being more evident in cheese packaged in LBMD/starch-based bioplastic films than in the one packaged in PA/PE. However, this was not observed by any decrease of pH values ($\text{pH} = 6$). After 21 days, the evaluators described the cheese packaged in LBMD/starch-based bioplastic films as having no perceived acidity. In

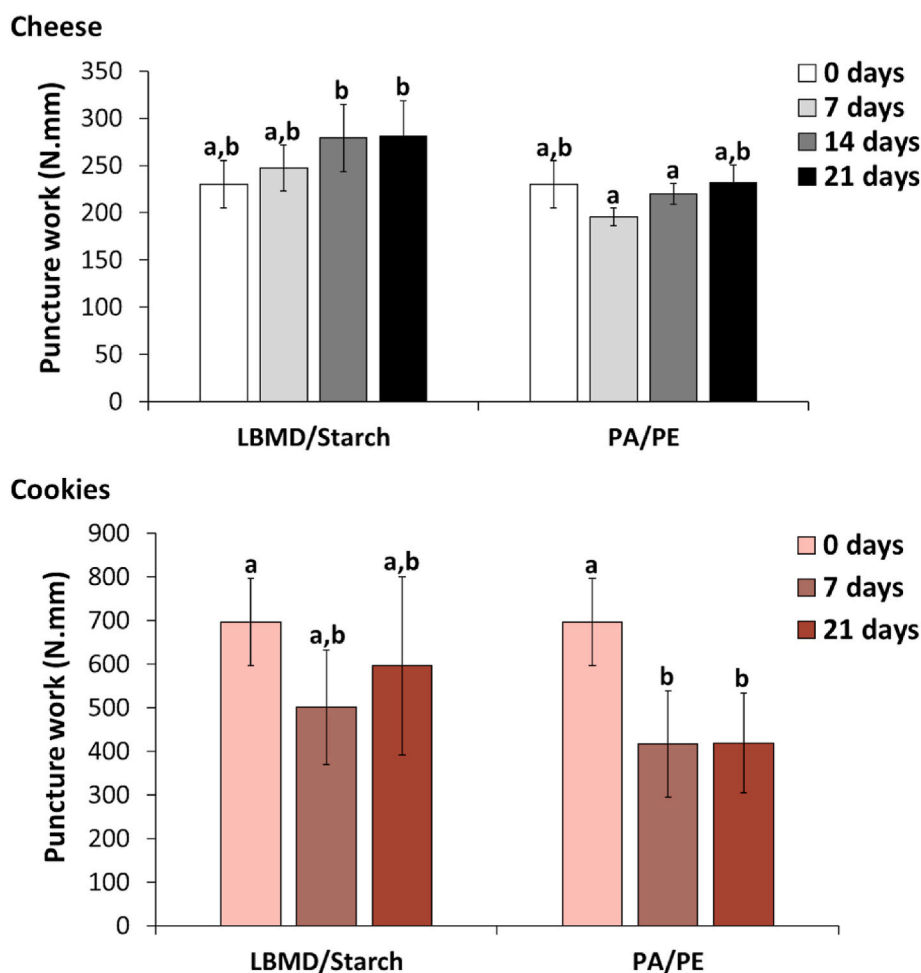


Fig. 5. Probe puncture work on cheese packaged with LBMD/starch-based bioplastics and non-biodegradable PA/PE-based plastic after 7, 14, and 21 days, and on oat cookies after 7, and 21 days. Different letters represent values that are significantly different ($p < 0.05$).

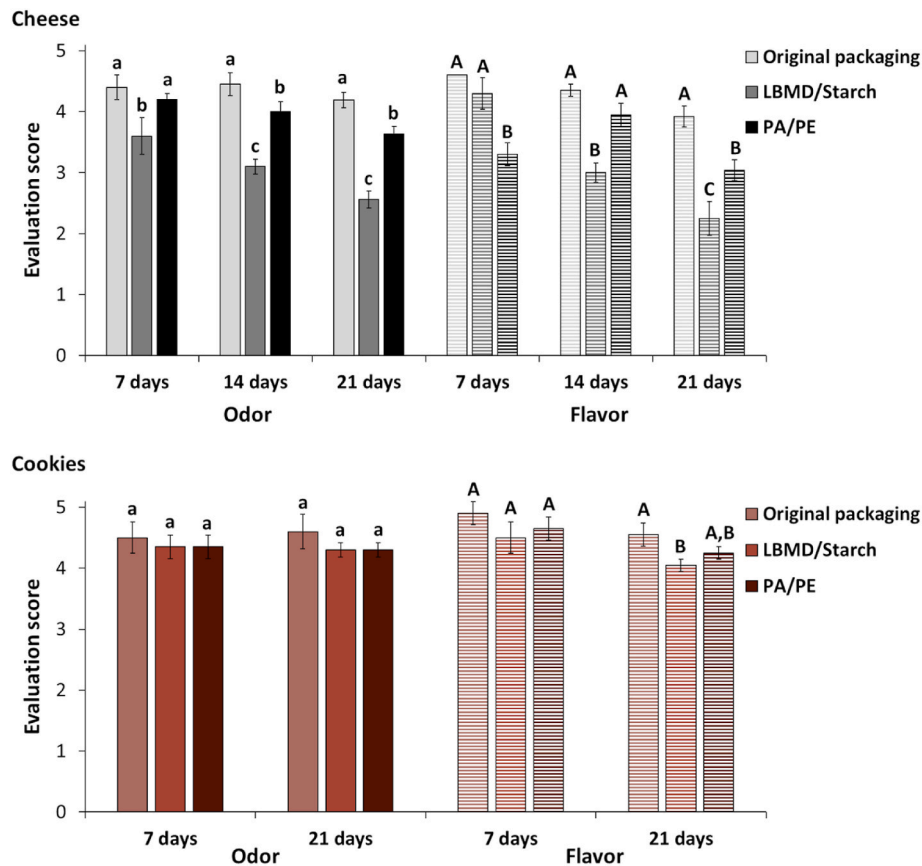


Fig. 6. Odor and flavor evaluation of cheese after 7, 14, and 21 days packaged with original packaging, LBMD/starch-based bioplastic films, and non-biodegradable PA/PE-based plastic; and oat cookies after 7, and 21 days. Evaluation scale: 1, poor quality; 5, excellent quality. Different letters represent significantly different values within each sampling time ($p < 0.05$).

addition, the hardness of the cheese packaged in LBMD/starch-based bioplastic films were also perceived to increase, described as becoming dried. This was in accordance with the weight loss measured for these samples (Fig. 4). After 14 days, some assessors noticed some rancid notes in the cheese packaged in LBMD/starch-based bioplastic films, in accordance with the decrease of the rate given by the evaluators to the odor after 14 days (Fig. 6).

3.2.5. Volatile compounds analysis

Furthermore, during storage time, the development of volatile compounds was evaluated (Table 4). Along the storage time, both cheese samples packaged in LBMD/starch-based bioplastic film and in PA/PE-based plastic showed an increased amount of volatile compounds. After 21 days, the volatile compounds increased in 182% for cheese packaged in bioplastics whereas increased in 130% for the one packaged in PA/PE-based plastic. In both samples, this increase was verified more in the group of aldehydes and ketones, when compared with alcohols and acids. However, the increase rate was different for the two packages. In cheese packaged in LBMD/starch-based bioplastic films, there was an initially large increase in volatile compounds (43%). From 7 to 14 days, this increase tended to be less pronounced (30%), increasing up to 52% from 14 to 21 days. Contrarily, in the cheese packaged in PA/PE-based plastic it was not observed such large increase in the first 14 days (7% in the first 7 days and 23% from day 7–14). However, the increase of volatile compounds content after this period was higher than that observed in bioplastics (76%).

The aldehydes with the largest increase in cheese in contact with plastic material were benzaldehyde (11 and 2 times in LBMD/starch-based bioplastic films and PA/PE-based plastic, respectively), 3-methylbutanal (8 times and 5 times), phenylacetaldehyde (7 and 3 times), and

nonanal (5 and 2 times), when compared to the cheese at day 0. The ketones with the largest increase were 3-hydroxybutanone (32 times in both samples) and 2,3-butanedione (9 and 5 times). These aldehydes and ketones are reported to result from lipid oxidation (Aparicio-Ruiz et al., 2020; Kochhar, 1996). In fact, the odor thresholds of the compounds related to rancidity, such as nonanal (10 $\mu\text{g}/\text{kg}$ of cheese) (Wang et al., 2021), and 3-hydroxybutanone (850 $\mu\text{g}/\text{kg}$ of cheese) (Wang et al., 2021) are much lower than the amount of these compounds, found in all samples even in sample at day 0 (3860 $\mu\text{g}/\text{kg}$ of cheese and 1240 $\mu\text{g}/\text{kg}$ of cheese, respectively). These compounds can contribute to the development of perceived odors and flavors noticed during the evaluation. Furthermore, in the case of LBMD/starch-based bioplastic films, a higher increase in 3-methylbutanal, phenylacetaldehyde, and benzaldehyde was observed which may result from Maillard reactions promoted by decarboxylation and oxidative deamination of amino acids (Rocha et al., 2021). These compounds can provide floral and malty aromas (Whetstone et al., 2006; Zehentbauer & Reineccius, 2002). For example, benzaldehyde has a threshold of 325 $\mu\text{g}/\text{kg}$ of cheese (Wang et al., 2021), lower than the amount found in all samples (Table 4), allowing to infer its contribution with almond notes (Peinado et al., 2004; Wang et al., 2021).

3.3. Packaging of oat cookies

3.3.1. General physicochemical analysis, texture analysis and odor and flavor evaluation

Oat cookies stored in LBMD/starch-based materials maintained the original appearance (day 0) of over 21 days, similar to the ones packaged with PA/PE-based packaging (Fig. 3B). The lightness (L^*) and red-green (a^*) chromatic parameters of oat cookies did not change over time

Table 4

Volatile compounds identified on cheese packaged with LBMD/starch-based bioplastic films and non-biodegradable PA/PE-based, after 7, 14, and 21 days of storage. The values are expressed in μg of ethyl heptanoate eq./g of cheese.

Compounds	Rt ^a (min)	LBMD/Starch				PA/PE		
		Day 0	Day 7	Day 14	Day 21	Day 7	Day 14	Day 21
Aldehydes								
2-Methylpropanal	3.216	–	0.68 ± 0.01 ^b	0.84 ± 0.01 ^{b,c}	1.44 ± 0.09 ^d	0.37 ± 0.01 ^a	0.68 ± 0.10 ^b	0.96 ± 0.05 ^c
2-Methylbutanal	5.735	–	–	0.78 ± 0.04 ^a	1.42 ± 0.13 ^b	–	0.59 ± 0.14 ^a	0.98 ± 0.32 ^{a,b}
3-Methylbutanal	5.946	1.30 ± 0.18 ^a	4.61 ± 0.33 ^{a,b,c}	5.90 ± 0.33 ^c	10.34 ± 1.54 ^d	2.35 ± 0.13 ^{a,b}	4.74 ± 0.62 ^{b,c}	6.66 ± 1.40 ^c
Hexanal	17.995	–	2.76 ± 0.33 ^b	2.42 ± 0.10 ^b	3.30 ± 0.47 ^b	–	–	1.22 ± 0.28 ^a
Octanal	33.304	2.12 ± 0.20 ^{a,b}	2.16 ± 0.66 ^{a,b}	3.61 ± 0.27 ^b	4.46 ± 1.43 ^b	–	0.52 ± 0.35 ^a	2.07 ± 0.31 ^{a,b}
Nonanal	36.824	3.86 ± 1.20 ^a	8.96 ± 2.46 ^{a,b,c}	16.07 ± 1.36 ^{b,c}	19.03 ± 6.74 ^c	1.65 ± 0.48 ^a	3.60 ± 0.59 ^a	6.49 ± 0.33 ^{a,b}
Decanal	39.510	0.31 ± 0.44 ^a	0.52 ± 0.08 ^a	0.28 ± 0.39 ^a	0.81 ± 0.11 ^a	–	0.54 ± 0.76 ^a	0.21 ± 0.30 ^a
Benzaldehyde	40.236	0.66 ± 0.06 ^a	1.54 ± 0.37 ^a	3.23 ± 0.27 ^b	7.42 ± 0.95 ^c	0.71 ± 0.05 ^a	1.35 ± 0.25 ^a	1.25 ± 0.01 ^a
Phenylacetaldehyde	42.793	0.59 ± 0.24 ^a	2.49 ± 0.32 ^{a,b}	2.85 ± 0.26 ^{a,b}	4.18 ± 1.35 ^b	0.75 ± 0.06 ^a	2.16 ± 0.47 ^{a,b}	1.66 ± 0.31 ^a
Total aldehydes		8.84	23.72	35.98	52.40	5.82	14.17	21.50
Ketones								
Acetone	3.286	13.89 ± 4.02 ^{a,b}	3.66 ± 0.37 ^{b,c}	1.15 ± 0.17 ^c	1.68 ± 0.46 ^c	13.79 ± 3.03 ^{a,b}	12.52 ± 3.84 ^{a,b}	16.78 ± 2.74 ^a
2-Butanone	5.340	17.04 ± 0.49 ^a	4.85 ± 0.69 ^b	1.74 ± 0.27 ^b	1.64 ± 0.47 ^b	18.69 ± 0.00 ^a	20.36 ± 3.26 ^a	26.51 ± 5.42 ^a
2-Pentanone	8.956	1.46 ± 0.11 ^{a,b,c}	1.10 ± 0.07 ^{a,b,c}	–	0.50 ± 0.71 ^{a,b}	1.96 ± 0.48 ^{b,c,d}	2.73 ± 0.48 ^{c,d}	3.37 ± 0.69 ^d
2,3-Butanedione	9.679	5.94 ± 0.29 ^a	20.38 ± 1.32 ^a	28.02 ± 0.59 ^{a,b}	54.43 ± 19.94	15.53 ± 2.93 ^a	19.41 ± 1.06 ^a	28.52 ± 3.32 ^{a,b}
2-Heptanone	27.033	3.22 ± 0.15 ^a	4.72 ± 0.95 ^{a,b,c}	3.66 ± 0.25 ^{a,b}	6.62 ± 0.53 ^c	4.02 ± 0.09 ^{a,b}	4.43 ± 0.62 ^{a,b,c}	5.85 ± 0.82 ^{b,c}
3-Hydroxybutanone	33.147	1.24 ± 0.04 ^a	28.01 ± 7.26 ^a	19.32 ± 7.18 ^a	41.23 ± 21.98	18.21 ± 0.52 ^a	21.89 ± 6.64 ^a	39.43 ± 24.50 ^a
2-Nonanone	36.665	2.13 ± 0.09 ^a	3.18 ± 0.68 ^a	3.88 ± 0.45 ^a	6.31 ± 0.55 ^b	3.25 ± 0.12 ^a	3.23 ± 0.87 ^a	3.61 ± 0.85 ^a
2-Undecanone	41.659	1.41 ± 0.04 ^a	1.81 ± 0.10 ^{a,b}	3.10 ± 0.72 ^{a,b}	3.75 ± 0.93 ^b	1.50 ± 0.07 ^a	1.62 ± 0.38 ^a	1.94 ± 0.61 ^{a,b}
Acetophenone	42.922	–	0.57 ± 0.02 ^{a,b}	1.08 ± 0.06 ^b	1.32 ± 0.58 ^b	–	0.92 ± 0.41 ^{a,b}	–
2-Tridecanone	45.632	0.39 ± 0.00 ^a	0.29 ± 0.41 ^a	0.67 ± 0.14 ^a	0.98 ± 0.06 ^a	–	0.43 ± 0.11 ^a	0.71 ± 0.53 ^a
Total ketones		46.72	68.58	62.60	118.45	76.95	87.54	126.72
Alcohols								
2-Butanol	12.913	21.07 ± 2.42 ^a	–	–	–	–	–	–
1-Butanol	24.466	–	1.53 ± 0.52 ^a	2.63 ± 0.33 ^b	3.05 ± 0.18 ^b	–	–	–
3-Methyl-3-buten-1-ol	31.559	0.96 ± 0.07 ^a	–	–	–	0.69 ± 0.10 ^{a,b}	0.52 ± 0.12 ^b	0.69 ± 0.22 ^{a,b}
1-Pentanol	31.674	1.06 ± 0.00 ^a	1.65 ± 0.26 ^a	2.23 ± 0.53 ^a	2.66 ± 0.75 ^a	1.12 ± 0.01 ^a	1.67 ± 0.18 ^a	1.95 ± 0.61 ^a
2-Heptanol	34.524	0.83 ± 0.05 ^a	–	–	–	–	–	–
3-Methyl-2-buten-1-ol	34.603	1.57 ± 0.01 ^a	1.02 ± 0.08 ^{a,b}	0.88 ± 0.05 ^b	1.08 ± 0.09 ^{a,b}	1.12 ± 0.06 ^{a,b}	1.05 ± 0.39 ^{a,b}	0.98 ± 0.22 ^{a,b}
1-Butoxy-2-propanol	35.149	–	1.13 ± 0.86 ^a	3.41 ± 0.55 ^b	3.21 ± 1.15 ^b	–	–	–
1-Hexanol	35.611	0.50 ± 0.02 ^a	3.09 ± 1.93 ^a	11.83 ± 1.83 ^{a,b}	16.17 ± 7.29 ^b	0.76 ± 0.05 ^a	1.07 ± 0.13 ^a	1.05 ± 0.35 ^a
2-Butoxyethanol	37.017	–	0.41 ± 0.58 ^a	2.37 ± 0.49 ^b	3.28 ± 2.01 ^{a,b}	–	–	–
1-Heptanol	38.399	0.45 ± 0.05 ^a	0.91 ± 0.26 ^{a,b}	1.85 ± 0.33 ^{a,b}	2.39 ± 1.00 ^b	–	0.32 ± 0.46 ^a	0.60 ± 0.01 ^a
2-Ethyl-1-hexanol	39.224	0.29 ± 0.41 ^a	2.55 ± 1.24 ^{a,b}	8.24 ± 1.09 ^b	9.35 ± 5.10 ^{a,b}	–	–	0.45 ± 0.64 ^a
1-Octanol	40.729	0.53 ± 0.22 ^a	0.99 ± 0.47 ^a	3.97 ± 0.73 ^a	4.73 ± 3.24 ^a	0.60 ± 0.09 ^a	0.76 ± 0.19 ^a	0.75 ± 0.30 ^a
Total alcohols		27.26	13.28	37.40	45.92	4.29	5.40	6.47
Acids								
Acetic acid	38.611	–	4.09 ± 1.35 ^{a,b}	8.62 ± 1.69 ^{b,c}	6.17 ± 2.39 ^{a,b,c}	1.15 ± 0.30 ^{a,b}	1.62 ± 0.56 ^{a,b}	12.95 ± 3.89 ^c
Butanoic acid	42.367	–	8.69 ± 1.88 ^a	9.44 ± 1.59 ^a	10.98 ± 2.85 ^a	–	–	23.18 ± 6.44 ^b
Total acids			12.78	18.06	17.15	1.15	1.62	36.13
Total		82.82	118.36	154.04	233.92	88.21	108.74	190.82

^a Rt – Retention time.

when packaged in LBMD/starch-based bioplastic films, similar to those packaged in PA/PE-based plastic, while a slight increase in the yellow-blue value (b^*) was observed when packaged in LBMD/starch-based bioplastic films (Table 1). Furthermore, the cookies texture did not significantly change during storage when packaged in LBMD/starch-based bioplastic films (Fig. 5), whereas when packaged in the PA/PE-based plastic, the firmness of the cookies decreased over time. The low water vapor barrier properties of bioplastic films can facilitate the diffusion of water molecules accumulated in the packaging when moisture changes happen during storage, thus minimizing the cookies water uptake and maintaining their crispness. In an opposite way, materials with low WVTR retain water molecules inside the packaging, which can be absorbed by the food. Cookies flavor and odor were evaluated by a semi-trained lab panel. Other attributes, such as hardness, sweetness, and rancidity, were also qualitatively evaluated by the panelists. According to the evaluation by the lab panel, the cookies packaged in LBMD/starch-based bioplastic films maintained their texture, sweetness, odor, and taste during the entire storage period, similar to the ones packaged in PA/PE-based plastic or cookies at day 0, without showing rancidity development (Fig. 6).

3.3.2. Volatile compounds analysis

After 21 days of storage, the volatile compounds increased 18% for

cookies packaged in LBMD/starch-based bioplastic films, significantly less than those packaged with PA/PE-based plastic that increased 120% (Table 5). The volatile compounds that most increased, in both packages, during storage were pentanal, hexanal, and heptanal (Table 5), indicating the occurrence of oxidative rancidity (Zbikowska et al., 2018). However, they were more evident for cookies packaged with PA/PE plastic. The lower increase of volatile compounds in cookies packaged in LBMD/starch-based bioplastic films may be due to the presence of phenolic compounds that prevent the lipids oxidation in cookies over time (Lopes et al., 2023). Nevertheless, the rancid odor was not detected by any of the assessors in cookies packaged neither with LBMD/starch-based bioplastic film nor PA/PE-based plastic. Hydroxyacetone and 3-hydroxybutanone increased in cookies packaged with PA/PE-based plastic after 21 days but were not observed when packaged with LBMD/starch-based bioplastic films. These compounds are reported to be formed during the production of cookies (Hofmann, 1999; Martins et al., 2000). It is possible that amine groups of protein present in LBMD/starch-based bioplastic films scavenge hydroxyketones via Maillard reaction, thus diminishing their presence in the cookies. It cannot be disclosed that this effect can also be related to the permeability of bioplastic films to these compounds.

Table 5

Volatile compounds identified on oat cookies packaged with LBMD/starch-based bioplastic films and non-biodegradable PA/PE-based plastic, after 7, and 21 days of storage. The values are expressed in μg of ethyl heptanoate eq./g of oat cookies.

Compounds	Rt ^a (min)	LBMD/starch			PA/PE	
		Day 0	Day 7	Day 21	Day 7	Day 21
Aldehydes						
2-Methylpropanal	3.208	2.36 ± 0.32 ^a	3.75 ± 0.78 ^a	4.72 ± 1.49 ^a	2.03 ± 0.01 ^a	4.85 ± 1.34 ^a
2-Methylbutanal	5.732	8.46 ± 1.04 ^{a,b}	4.72 ± 0.64 ^a	4.54 ± 1.10 ^a	6.37 ± 0.30 ^a	14.13 ± 3.99 ^b
3-Methylbutanal	5.939	8.43 ± 1.11 ^{a,b}	5.39 ± 0.67 ^a	4.99 ± 1.18 ^a	7.20 ± 0.38 ^a	16.04 ± 4.51 ^b
Pentanal	9.124	0.58 ± 0.29 ^a	1.19 ± 0.89 ^a	1.22 ± 0.76 ^a	–	13.05 ± 3.11 ^b
Hexanal	17.990	13.17 ± 2.91 ^a	35.99 ± 6.17 ^{a,b}	23.18 ± 6.82 ^a	17.10 ± 1.48 ^a	61.02 ± 15.20 ^b
Heptanal	27.314	1.70 ± 0.30 ^a	4.23 ± 0.77 ^{a,b}	3.57 ± 0.94 ^{a,b}	2.25 ± 0.16 ^a	5.61 ± 0.94 ^b
Octanal	33.300	0.95 ± 0.10 ^a	3.19 ± 0.78 ^{a,b}	3.51 ± 0.78 ^{a,b}	2.03 ± 0.02 ^a	5.28 ± 1.00 ^b
Nonanal	36.814	2.77 ± 0.74 ^a	10.40 ± 2.66 ^{b,c}	13.31 ± 2.15 ^c	4.17 ± 0.07 ^{a,b}	9.67 ± 1.57 ^{b,c}
Decanal	39.502	–	1.35 ± 0.48 ^a	1.27 ± 0.03 ^a	0.80 ± 0.15 ^a	0.81 ± 1.14 ^a
Benzaldehyde	40.225	3.20 ± 0.37 ^{a,b}	4.89 ± 0.79 ^{a,b}	6.13 ± 1.12 ^b	2.44 ± 0.03 ^a	5.70 ± 1.17 ^{a,b}
Total aldehydes		41.65	75.10	66.44	44.40	136.16
Ketones						
Acetone	3.275	5.18 ± 1.37 ^a	7.16 ± 0.35 ^a	9.56 ± 1.34 ^a	3.06 ± 0.28 ^a	8.16 ± 3.05 ^a
Butanone	5.330	3.26 ± 0.51 ^a	3.42 ± 0.49 ^a	3.62 ± 0.97 ^a	2.37 ± 0.24 ^a	6.09 ± 1.68 ^a
Butanedione	9.671	8.97 ± 0.44 ^a	31.58 ± 6.00 ^{a,b}	37.43 ± 12.16 ^b	8.84 ± 0.58 ^a	22.12 ± 5.82 ^{a,b}
3-Hexanone	16.687	3.66 ± 0.15 ^a	4.62 ± 0.74 ^a	3.05 ± 1.20 ^a	–	–
3-Hydroxybutanone	33.093	4.07 ± 0.35 ^a	7.95 ± 1.72 ^a	5.46 ± 2.16 ^a	4.25 ± 0.60 ^a	10.38 ± 2.38 ^a
Hydroxyacetone	33.797	53.00 ± 11.68 ^a	46.71 ± 12.73 ^a	27.45 ± 14.53 ^a	47.40 ± 9.59 ^a	89.90 ± 25.05 ^a
Total ketones		119.80	176.55	153.01	110.32	272.80
Alcohols						
1-Methoxy-2-propanol	22.405	6.53 ± 0.48 ^{a,b}	2.20 ± 0.11 ^c	–	3.54 ± 1.75 ^{b,c}	7.02 ± 0.03 ^a
1-Pentanol	31.668	1.31 ± 0.10 ^a	3.21 ± 0.61 ^{a,b}	2.59 ± 0.72 ^{a,b}	1.35 ± 0.09 ^a	5.51 ± 1.36 ^b
1-Hexanol	35.598	–	1.49 ± 0.16 ^a	1.26 ± 0.35 ^a	–	–
1-Octen-3-ol	38.291	0.96 ± 0.08 ^a	1.79 ± 0.41 ^a	2.88 ± 0.80 ^a	0.92 ± 0.04 ^a	3.56 ± 1.56 ^a
1-Octanol	40.717	–	0.89 ± 0.21 ^{a,b}	1.23 ± 0.32 ^{b,c}	0.77 ± 0.01 ^{a,b}	2.03 ± 0.42 ^c
Total alcohols		8.80	9.57	7.96	6.59	18.12
Acids						
Acetic acid	38.555	28.10 ± 0.22 ^a	6.11 ± 0.02 ^c	5.63 ± 0.06 ^c	5.27 ± 0.55 ^c	9.25 ± 0.13 ^b
Total acids		28.10	6.11	5.63	5.27	9.25
Total		198.35	267.33	233.04	166.58	436.33

^a Rt – Retention time.

4. Conclusions

Blending LBMD with starch allowed to develop transparent and yellow bioplastic films with higher rigidity, hydrophobicity, and gas barrier properties than the bioplastic films prepared only with LBMD or starch. The initial food quality of cheese were protected until 14 days of storage in LBMD/starch-based bioplastic films at 4 °C and 80% relative humidity. After this period, oxidative reactions were sensorially perceived through odor and flavor, although not in PA/PE-based plastic. Moreover, the bioplastic packaging allowed cheese dehydration (*ca.* 10%), which can be exploited as a technological approach in cheese ripening, which was not evident when PA/PE-based plastic was used. Concerning oat cookies, after 21 days, LBMD/starch-based bioplastic films seemed to be suitable to preserve oat cookies at 20 °C and 45% relative humidity, as these bioplastic films prevented the perceived oxidative rancidity of fatty cookies, contrary to PA/PE-based plastic. The bioplastic films showed to be useful to mitigate oxidative reactions as well as to scavenge the formation of volatile compounds in this roasted derived food, such as hydroxyketones. Thus, blending LBMD with starch allowed to obtain active bioplastic films with water absorption, protection against UV radiation, volatile compounds scavenging capacity, and antioxidant activity, capable of preserving fatty foodstuffs while promoting new characteristics and flavors to them.

Author statement

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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

No data was used for the research described in the article.

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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.2023.109322>.

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