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Physicochemical Characterization and Biodegradability of Nanostructured Chitosan-Based Films Reinforced with Orange Waste

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Abstract

The valorization of agricultural by-products through their integration into biodegradable materials represents a promising approach for sustainable food preservation. In this study, nanostructured chitosan/polyvinyl alcohol (PVA)/orange peel-bagasse waste (OPB) (0.125%, 0.25%, and 0.5% OPB) films were developed and characterized for their physicochemical, mechanical, and biodegradation properties. Scanning electron microscopy and confocal laser scanning microscopy revealed that OPB concentration influenced structural homogeneity. Attenuated total reflectance-Fourier transform infrared spectroscopy (ATR-FTIR) revealed possible molecular interactions among components through hydrogen bonding (peaks at 1570, 1416 cm⁻¹, and 1020 cm⁻¹) and imine (C = N) formation (broadening of the peak at 1425 cm⁻¹). As OPB increased, water vapor diffusion and film rigidity increased, while elongation at break decreased. After composting, weight loss was 93.7% and 100% for the chitosan and PVA films, respectively. For the nanostructured films, weight loss was between 94.7% (30PVA/0.5OPB) and 99.7% (30PVA/0.125OPB). Regarding ATR-FTIR of the blends, the intensity of the peaks located between 3625 and 3005 cm⁻¹, at 2919 cm⁻¹, at 1729 cm⁻¹, at 1621 cm⁻¹, at 1521 cm⁻¹, and between 1160 and 885 cm⁻¹, corresponding to the OPB functional groups, decreased. These results demonstrate that incorporating citrus waste enhances biodegradability and provides films barrier properties suitable for fresh produce preservation.

Keywords: biodegradable films; chitosan blends; orange waste; biodegradation; food packaging



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1. Introduction

The growing volume of agricultural and food processing residues poses a significant environmental challenge in this century. Large quantities of lignocellulosic by-products are discarded annually, contributing to waste accumulation and resource inefficiency. The transition toward a circular economy requires the development of value-added materials that convert these residues into functional products. Among such approaches, biodegradable polymer composites have attracted growing attention due to their capacity to reduce

plastic pollution while providing suitable physicochemical and mechanical properties for packaging and preservation applications. Natural polymers such as chitosan (CS), when blended with synthetic biodegradable matrices like polyvinyl alcohol, offer a promising platform for developing environmentally friendly materials. In this context, citrus processing industries produce considerable amounts of peel and bagasse residues rich in pectin, cellulose, and polyphenolic compounds, which can be reused as natural fillers to improve film performance and accelerate biodegradation.

Currently, new alternatives for edible films with natural products and reuse of agricultural products are subjects of special interest. A natural polysaccharide such as chitosan has been widely recognized for its antimicrobial properties, non-toxicity, and biodegradability [1,2]. It has been used as a film for food packaging applications [3,4]. To improve the performance of biodegradable films, polymer blending is a good alternative [5]. The blends of PVA (polyvinyl alcohol) and chitosan have shown higher miscibility, as PVA is a thermoplastic biodegradable polymer [6,7]. Moreover, the incorporation of nanostructures into edible films is a new strategy to improve their performance [8]. Chitosan nanoparticles incorporated into edible films have been proven to increase the shelf life of horticultural products such as mango, tomato, and bell peppers, among others [9–11].

Although chitosan/PVA blends produce biodegradable films, their strength, functionality, and disintegration rate could be increased by adding cellulose-rich materials, such as agricultural by-products. In particular, the citrus fruit industry generates many by-products such as peels and bagasse [12]. In Mexico, approximately 50% of a fruit's weight is converted into agro-industrial waste, amounting to 800,000 t per year [13,14]. One way to revalorize this product is to use it as a raw material for biofuels, essential oil, pectin extraction, and animal feed, and most recently, it has been incorporated into biodegradable films [6,15]. Yaradoddi et al. [16] pulverized orange peel and added hydrochloric acid to extract pectin and glycerol as a plasticizer to form casting films. The authors concluded that these films could have important applications in the industry because orange peel is a naturally derived resource with no harmful constituents. The cellulosic or pectin fibers give the film strength, ensuring biodegradability and lower thermal disintegration, as assessed by thermogravimetric analysis (TGA). Yi et al. [17] prepared casting films from PVA films and cellulose extracted from orange peel using an alkaline treatment with sodium hydroxide. Using FTIR, they found that intra- and intermolecular hydrogen bonds were formed between PVA and cellulose. As the cellulose content increased, the tensile strength and elongation at break decreased, while Young's modulus increased. Rathinavel et al. [18] incorporated orange peel powder (5 to 20 wt. %) into PVA matrix biocomposite films by casting. FTIR analysis showed interactions between orange peel powder and PVA functional groups. The films were thermally stable up to 350 °C, as measured by TGA, and the tensile strength increased from 6.20 MPa to 7.80 MPa after orange peel powder was incorporated. The film's thickness was suitable for edible film production. Sánchez-Orozco et al. [19] also prepared sodium alginate–orange peel edible films using glycerol as a plasticizer. The films were characterized in terms of their structural, thermal, mechanical, and barrier properties. FTIR spectra revealed interactions between orange peel and sodium alginate functional groups. Regarding the mechanical properties, the tensile strength increased from 0.92 to 7.39 MPa, and elongation at break decreased from 20.76 to 2.67% after the orange peel was incorporated, making the orange peelalginate-glycerol films a good alternative for food packaging. Taghavi et al. [20] studied the effect of incorporating different concentrations (0, 3, 6, 9, 12, and 15%) of orange peel powder into gelatin films. The moisture content and water vapor permeability of the films increased as the orange peel content increased. Scanning electron microscopy (SEM) images showed that as orange peel powder increased, the film was not homogeneous and had white spots due to the immiscibility of the orange

particles. Changes in color were also observed; however, opacity increased. Higher tensile strength and lower elongation at break were observed after orange peel incorporation. Terzioğlu et al. [6] incorporated orange peel powder (0.25–1.25% *w/w*) into chitosan/PVA casting films. FTIR revealed an interaction between the orange peel and the chitosan/PVA functional groups. The thermal stability (TGA) and water vapor permeability were increased due to the incorporation of the orange peel. On the other hand, the transparency of the film was decreased as measured by UV-vis spectroscopy. Also, hydrophobicity and oxygen transmission rate decreased, increasing the antioxidant activity of the films. They found that for a concentration of 2 wt% pectin, there was a uniform dispersion of curcumin nanoparticles in the film, and as the concentration of nanoparticles increased, the films were rough and porous. FTIR results showed good interaction between the pectin and chitosan functional groups. In previous research work, nanostructured chitosan-PVA films were developed for blueberry preservation [21]. FTIR analysis showed interactions between chitosan and PVA functional groups. The incorporation of chitosan nanoparticles into these composite films increased thermal stability and Young's modulus. Therefore, the present study aims to develop and characterize nanostructured chitosan/polyvinyl alcohol (CS/PVA) films reinforced with orange peel–bagasse (OPB) waste as a sustainable material for food packaging applications. It is hypothesized that incorporating lignocellulosic residues from citrus waste enhances film biodegradability while modulating mechanical and barrier properties through hydrogen bonding and electrostatic interactions within the polymeric matrix. The novelty of this work lies in integrating agricultural orange waste into chitosan/PVA nanocomposites, combining environmental valorization with improved material functionality for fresh produce preservation. This approach advances circular economy strategies by transforming agro-industrial by-products into biodegradable films with potential use in sustainable food preservation systems.

2. Materials and Methods

Two chitosan (CS) solutions were prepared. For the nanoparticles' aqueous phase, medium molecular weight CS (deacetylation degree of 75–85%) was purchased from Sigma Aldrich® (St. Louis, MO, USA). For the film-forming solution, high molecular weight CS (91% deacetylation degree) was bought from América Alimentos (Zapopan, Jal., Mexico). Glacial acetic acid, acquired from Fermont Chemicals Inc. (Monterrey, Mexico), and distilled water were used to prepare the aqueous phase. For the organic phase, ethanol was purchased from JT Backer (Phillipsburg, NJ, USA), and Tween 20 was supplied by Hycel (Zapopan, Jal., Mexico). The PVA solution was prepared using distilled water and PVA acquired from Sigma Aldrich®, with a molecular weight of 3000–23,000 (Mw) and 87–89% hydrolyzed. For film formation, glycerol from JT Backer was used as a plasticizer and citric acid from Fermont was used as a crosslinker. The orange peel flour and bagasse (OPB) were donated by Centro de Innovación e Integración de Tecnologías Avanzadas del Instituto Politécnico Nacional (Veracruz, Mexico).

The nanoprecipitation or solvent displacement method was used for chitosan nanoparticle (NP) preparation [22]. Two phases were prepared: an aqueous phase containing CS (0.05% *w/v*) dissolved in acetic acid (1% *v/v*) and distilled water (pH 5.6), and an organic phase, in which 40 mL of ethanol and 10 µL of Tween 20 were mixed and kept under stirring. After that, 2.5 mL of the aqueous phase was added dropwise to the organic phase with the aid of a peristaltic pump (MasterFlex, Vernon Hills, IL, USA) and maintained under stirring for 10 min. The morphology of the obtained NPs was characterized by transmission electron microscopy (TEM) using a JEOL microscope (JEM 2010, Tokyo, Japan) at an acceleration voltage of 200 kV. Particle size distribution and Z potential were mea-

sured by dynamic light scattering (DLS) with a Zetasizer Nano-ZS90 instrument (Malvern Instruments, Worcestershire, UK).

The film-forming solution was prepared by dissolving the CS (1% *w/v*) in a mixture of acetic acid (1% *v/v*) and distilled water. A PVA (5% *w/v*) solution, which was previously dissolved in distilled water at 60 °C, was then added, along with the NPs. This mixture was stirred for 30 min using a homogenizer (Virtis, Gardiner, NY, USA) at 10,000 rpm. After that, citric acid (10%) was added, and the solution was stirred for an additional 10 min. The orange peel–bagasse blend (50/50) (OPB) was then added, and the solution was heated at 50 °C for 1 h (the OPB was previously oven-dried at 60 °C for 24 h). After cooling to room temperature, glycerol was added, and the solution was stirred for 30 min. Finally, 7 mL of the film-forming solution was added to a silicon mold and oven-dried (Binder, TUT, Germany) at 40 °C for 5 h to prepare the films by casting. The evaluated formulations are shown in Table 1.

Table 1. Formulations evaluated for the films.

Formulation	Components (%)					
	PVA	NP	OPB	CS	Gly	CA
CSNP	-	33	-	67	-	-
PVA	100	-	-	-	-	-
30PVA/0.125OPB	30	33	0.125	34.875	1	1
30PVA/0.25OPB	30	33	0.250	34.750	1	1
30PVA/0.5OPB	30	33	0.500	34.500	1	1
40PVA/0.125OPB	40	33	0.125	24.875	1	1
40PVA/0.25OPB	40	33	0.250	24.750	1	1
40PVA/0.5OPB	40	33	0.500	24.500	1	1

CSNP = chitosan nanoparticles + chitosan solution (1%), PVA = polyvinyl alcohol, NPs = chitosan nanoparticles, OPB = orange peel–bagasse, CS = chitosan (1%), Gly = glycerol, CA = citric acid.

An environmental scanning electron microscope (Carl Zeiss, EVO LS10, Germany) was used to observe the cross-section of cryogenic fractured samples. The acceleration voltage was 20 kV. Moreover, surface morphology was observed using confocal laser scanning microscopy or CLSM (Carl Zeiss, LSM 800, Oberkochen, Germany) coupled to a color AxioCam HD (Carl Zeiss, Model 305, Oberkochen, Germany). ZEN (Zeiss efficient navigation) software version 2.6 Blue edition was used. The micrographs were acquired with a 20X apochromatic objective and a numerical aperture of 0.8. Autofluorescence was identified using the “lambda mode” tool, and 488 nm and 560 nm lasers were used at 4% excitation with a pinhole aperture equivalent to 1 airy unit (AU).

ATR-FTIR (attenuated total reflectance-Fourier transform infrared) spectroscopy (Thermo Scientific iS5 Nicolet, Thermo Fisher Scientific, Madison, WI, USA) was used to evaluate the functional groups and their interaction. Spectra were recorded from 4000 to 400 cm^{-1} at a resolution of 4 cm^{-1} over 24 scans.

Physical properties such as moisture content (MC) and water vapor diffusion coefficient (WVDC) were determined according to the methodology of Correa-Pacheco et al. [21]. For MC, square pieces of the films (2 × 2 cm) were cut and weighed. They were then placed in a vacuum oven (Yamato, model ADP200C, San Jose, CA, USA) at 40 ± 1 °C for 24 h. After that, the samples were weighed, and the MC was calculated as $\text{MC} (\%) = (w_i - w_d)/w_i \times 100$, with w_i and w_d representing the initial and final film weights after vacuum-drying, respectively. The photoacoustic technique was employed to determine WVDC. The film was placed between a quartz window and an aluminum foil in the photoacoustic cell and irradiated on one side using a Tungsten-Halogen lamp (Oriel Corporation 64,655, New York, NY, USA) with a modulated light beam (17 Hz, 250 W).

The other side was exposed to controlled relative humidity (RH) using distilled water, which was diffused through the sample. The generated thermal waves caused changes in pressure being sensed by an electret microphone coupled to the PA cell and were recorded as amplitude and phase as functions of time in a lock-in amplifier (Stanford Research Systems SR836 DSP, Sunnyvale, CA, USA). The following equation was used to calculate water diffusion: $WVDC = l^2/2t$, where l is the film thickness and t is the time for water molecules to diffuse through the film [22]. Measurements were made in triplicate.

Mechanical properties were evaluated using an Instron testing machine (model 3345, Norwood, MA, USA) according to ASTM D638-03. All testing was performed at a speed of 5 mm/min, room temperature (28 ± 1 °C), and with a 1 kN load cell. Prior to measurement, samples were conditioned for 48 h at room temperature and 40% RH for 48 h. Ten specimens were tested.

The standard used to assess degradation was ASTM D6400. The test was performed on the films cut into pieces (2×2 cm). The films were buried in a homemade compost at 5 cm depth in ventilated commercial plastic boxes at room temperature (40 ± 2 °C). Films were kept for 1 month, and samples were collected from the soil after one week, two weeks, and four weeks of composting. Five replicates were made. Soil pH, conductivity (Thermo Scientific potentiometer, Madison, WI, USA), and water activity (Sagaon Tech S.A. de C.V., model WA-160A, Qro., Querétaro, Mexico) of the substrate in which the films were composted was monitored before and after composting. Changes in the microbial composition of the compost were evaluated by thermophilic bacteria, mesophilic bacteria, and fungi and yeast count according to NOM-092-SSA1-1994. After composting, the samples were washed to remove soil and impurities and weighed to determine the percentage of weight loss. CLSM and ATR-FTIR were also used to follow film degradation.

The data was analyzed using InfoStat software (Version 2020). A one-way analysis of variance (ANOVA) and Tukey's test and the LSD test ($p < 0.05$) were performed.

3. Results

3.1. Chitosan NPs TEM, DLS, and Z Potential Characterization

Figure 1 shows the results from the TEM observation and the particle size distribution by DLS for the chitosan NPs. The TEM micrograph in Figure 1a reveals spherical NPs. As shown in Figure 1b, a bimodal distribution was observed for the chitosan NPs. The smallest population had a size between 4 and 35 nm (78.7%) with a mean peak diameter of 13.37 nm. The diameter for the largest population ranged from 300 to 3500 nm (21.3%), which is in agreement with previously published works [21,22]. The presence of aggregates could be associated with this larger particle size population [23]. Particle aggregation can increase microstructural heterogeneity causing phase separation. It can also act as stress concentration points, affecting the mechanical properties. A Z potential value of 18.5 mV was measured. This value indicates that although the chitosan solution is relatively stable, particles may still aggregate over time or in response to variations in pH or ionic strength [24]. The obtained value was lower than the ± 30 mV often reported in the literature for stable, monodisperse solutions [25].

Although the pH was kept at 5.6 to assure chitosan solubility and stability during experiments, possible reasons for aggregation can be related to the following [26]: (1) Charge interactions. It can be assessed by Z potential value. If the Z potential is low, as found in this work, the electrostatic interaction between particles decreases with subsequent aggregation; (2) polymer chain entanglement due to intermolecular hydrogen bonds of the hydroxyl and amino groups between the different components of the films causing aggregation; and (3) additional speed of chitosan through the peristaltic pump affects the diffusion between solvent and nonsolvent and can cause aggregation [27–29].

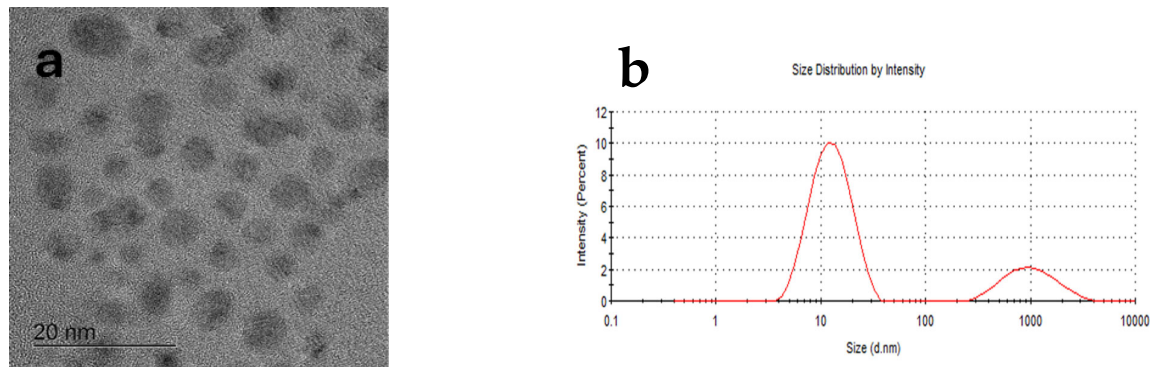


Figure 1. Chitosan NPs: (a) TEM micrograph and (b) DLS.

3.2. SEM and CLSM Film Characterization

Figure 2 shows the SEM of the OPB powder and films cross section micrographs of the CSNP, PVA, and polymer blend films. The OPB particles showed various shapes and heterogeneous particle size distribution as seen in Figure 2a. The thicknesses of the films measured from the images were the following: $58.49 \pm 0.19 \mu\text{m}$ (CSNP), $121.7 \pm 1.44 \mu\text{m}$ (PVA), $65.13 \pm 2.34 \mu\text{m}$ (30PVA/0.125OPB), $79.58 \pm 2.41 \mu\text{m}$ (30PVA/0.25OPB), $79.34 \pm 3.02 \mu\text{m}$ (30PVA/0.5OPB), $100.33 \pm 5.83 \mu\text{m}$ (40PVA/0.125OPB), $106.85 \pm 1.62 \mu\text{m}$ (40PVA/0.25OPB), and $109.27 \pm 7.61 \mu\text{m}$ (40PVA/0.5OPB). As PVA was added to the CSNP composition, the thickness increased. The cross-section of the CSNP film (Figure 2b) showed a rough structure. In contrast, the PVA film (Figure 2c) exhibited plastic deformation. The polymer film composites (Figure 2d–i) had similar structures, showing largely homogeneous mixing with minimal phase separation. On this line of research, Ayca et al. [30] and Terzioğlu et al. [6] observed the same morphology for CS/PVA blends. As pectin is an anionic polymer and chitosan is a cationic polymer, a complex network forms, resulting in improved physical properties, as observed by Zhou et al. [31] for pectin–chitosan films. However, small cracks and pores were seen, potentially due to disorganization of the CS/PVA matrix when OPB was incorporated or separated from the polymer matrix, especially for the 40% PVA films as observed in Figure 2g–i [6,20,32]. The observed microdefects (cracks and pores) are primarily attributed to two phenomena during film drying: (1) matrix disorganization caused by the incorporation of the OPB, a lignocellulosic filler, and (2) the hydrophilic nature of OPB, which can lead to slight separation from the polymeric matrix during solvent evaporation. The presence of cracks is related to internal stress between the CS/PVA blending polymer and OPB due to heating and cooling within the film [33,34]. Cracks will appear in the film if the stress is higher than the material's strength, especially at the interface between CS/PVA blend and the OPB. Pores can be formed during casting during solvent evaporation if the solvent evaporates unevenly [35]. Therefore, pores can form from processing rather than differences between phases' miscibility [34,36].

Figure 3 shows the CLSM of the films. For CSNP (Figure 3a), an uneven distribution of chitosan nanoparticles was seen with the presence of agglomerates. For the PVA film, a uniform surface was observed (Figure 3b). After OPB incorporation, a very light orange coloration due to OPB was found in the 30PVA/0.125OPB film (Figure 3c). A non-uniform distribution of orange waste was observed in the matrix, with small orange clusters or micro-separation of phases in the form of circles. A more homogeneous distribution was observed in the 30PVA/0.25OPB film (Figure 3d). In this case, a light orange color was still observed throughout the coating; however, small clusters of orange waste and small microphases were still found. As the OPB content increased to 30PVA/0.5OPB, the film

became darker with an orange coloration while the presence of microphases was lower, resulting in the film being more homogeneous (Figure 3e).

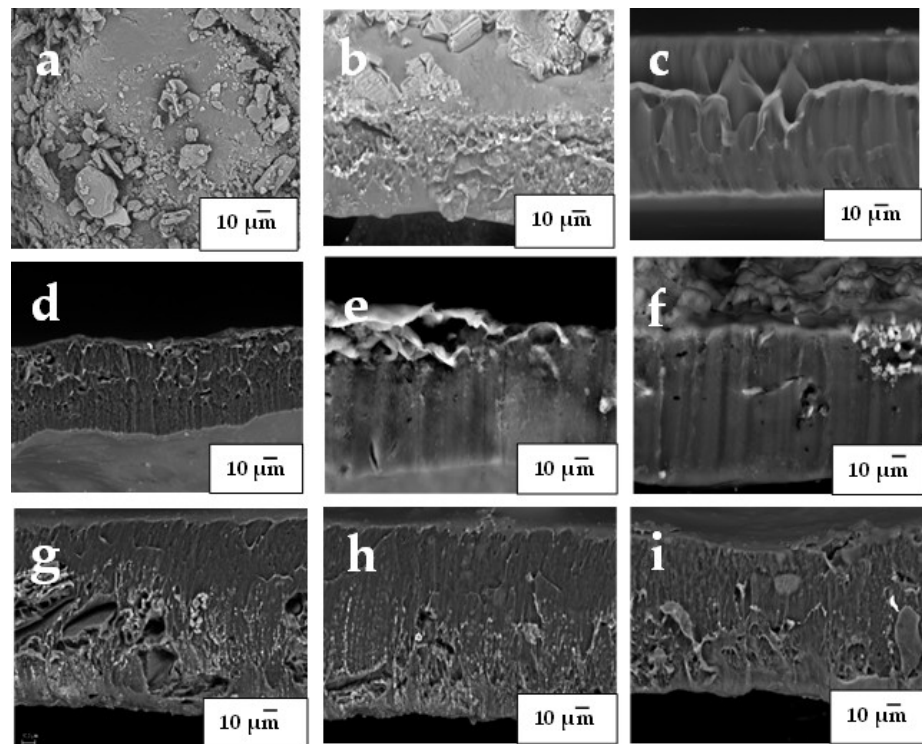


Figure 2. SEM of (a) OPB powder, and cross-sectional micrographs of the films, (b) CSNP, (c) PVA, (d) 30PVA/0.125OPB, (e) 30PVA/0.25OPB, (f) 30PVA/0.5OPB, (g) 40PVA/0.125OPB, (h) 40PVA/0.25OPB, and (i) 40PVA/0.5OPB (see Table 1).

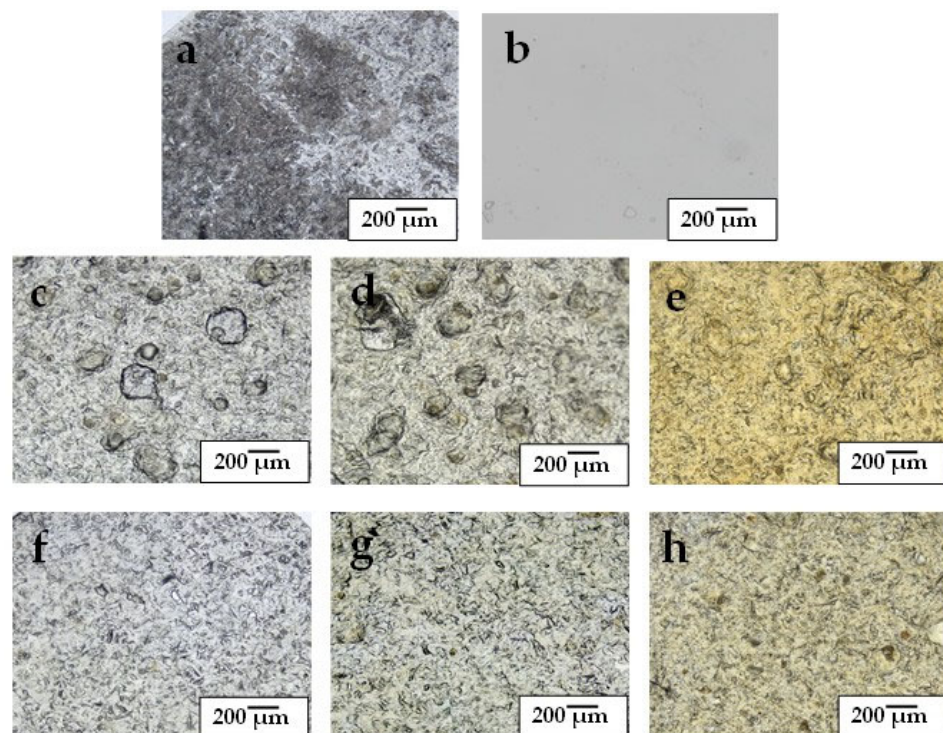


Figure 3. CLSM surface film micrographs of (a) CSNP, (b) PVA, (c) 30PVA/0.125OPB, (d) 30PVA/0.25OPB, (e) 30PVA/0.5OPB, (f) 40PVA/0.125OPB, (g) 40PVA/0.25OPB, (h) 40PVA/0.5OPB (see Table 1).

For films with 40% PVA (Figure 3f–h), less phase separation was observed. The PVA has many hydroxyl (-OH) groups in its chemical structure, which can interact with the pectin and lignin carboxyl and hydroxyl groups from OPB through hydrogen bond interactions. Although chitosan has -OH groups like PVA, it also has amino groups, which can interact through electrostatic interactions with the OPB -OH and (C = O) groups [6]. When more PVA is added, more hydrogen bonding interactions are favored due to -OH groups, and electrostatic interactions from protonated amino groups of chitosan are lowered within the polymer matrix [37,38]. Formation of these interactions was confirmed in the ATR-FTIR analysis, where the broad band between 3500 and 3000 cm^{-1} corresponding to the -OH stretching is broadened and increased in intensity in the blends, as indicative of strong hydrogen bonding interactions (see Section 3.3).

3.3. ATR-FTIR Film Characterization

Figure 4 shows the ATR-FTIR spectra of the OPB, neat polymers, and blends. For the OPB spectrum (Figure 4a), the main characteristic peaks appeared between 3625 and 3005 cm^{-1} , corresponding to the symmetric and asymmetric stretching of the -OH groups characteristic of lignin, pectin, cellulose, and polyphenolic compounds. Other peaks were observed at 2919 cm^{-1} for -CH stretching, at 1729 cm^{-1} for C = O stretching, and at 1621 cm^{-1} for the carboxylate ions (COO⁻) stretching of pectin. A peak at 1521 cm^{-1} is attributed to the C = C stretching vibration in aromatic rings, while a broad band between 1160 and 885 cm^{-1} corresponds to C–O–H vibration and out-of-plane and in-plane C–H bending for aromatics [6,39].

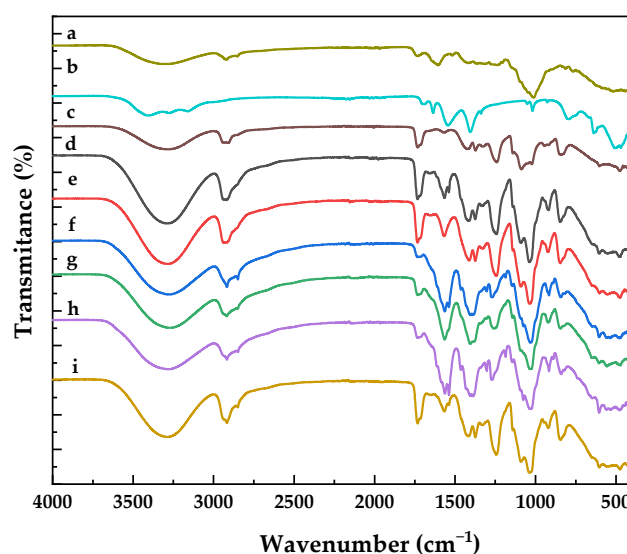


Figure 4. FTIR spectra of (a) OPB, (b) CSNP, (c) PVA, (d) 30PVA/0.125OPB, (e) 30PVA/0.25OPB, (f) 30PVA/0.5OPB, (g) 40PVA/0.125OPB, (h) 40PVA/0.25OPB, (i) 40PVA/0.5OPB films (see Table 1).

For CSNP (Figure 4b), the main peaks included a broad band between 3500 and 3000 cm^{-1} corresponding to -OH and -NH stretching, and a peak at 1700 cm^{-1} for C = O stretching. The bands at 1630 and at 1540 cm^{-1} are associated with the amide group's carbonyl bonds (amide I) and the amine group (amide II), respectively. Additionally, peaks were observed at 1411 cm^{-1} (related to -OH bending), 1020 cm^{-1} (C–O–C stretching), and 795 cm^{-1} (chitosan crystallization) [40–43].

The spectra for PVA (Figure 4c) showed several main bands. The band between 3600 and 3000 cm^{-1} corresponds to the -OH stretching. A band at 2924 cm^{-1} is associated with -CH₂ stretching, and a band at 1730 cm^{-1} is for the C = O in the PVA acetate units. Other peaks included one at 1426 cm^{-1} for the -CH vibration of the methyl group, one at

1145 cm^{-1} for the -CO group, another at 1080 cm^{-1} for the stretching vibration of the -CO acetate group, and finally, a peak at 835 cm^{-1} due to C-C bonding [42–44].

For the blends (Figure 4c–i), the peak at 3284 cm^{-1} broadened and increased in intensity, indicating interactions among CS, PVA, and OPB. Peaks at approximately 1570, 1416, and 1020 cm^{-1} also showed a doublet, which is correlated with hydrogen bonding or bonding of the -NH and -OH groups of the different blend components [43,45–48]. Finally, the peak at 1425 cm^{-1} ($\gamma\text{C-N}$) broadened, indicating the formation of N = C imine bonds from the interaction between chitosan and OPB [49].

3.4. MC, WVDC, and Mechanical Properties

Table 2 shows the results of MC, WVDC, and the mechanical properties of the films. The highest MC was for the CSNP film and the lowest for PVA. The blends generally had similar values, showing no statistical differences ($p < 0.05$) among them. Similar MC was found in previous work for CSNP ($35 \pm 0.07\%$) and PVA ($4 \pm 0.01\%$) films [21]. Zhou et al. [31] found a WC value 30% lower than for chitosan–pectin films and noted that as the pectin content increased, the WC decreased. This effect could be attributed to the interaction between the two polymers which decreased the binding of water to chitosan. Chitosan is a hydrophilic polymer due to the presence of hydroxyl and amine groups that can interact with water. While PVA is also hydrophilic, its synthetic nature limits its interaction with biological molecules because it is less hydrophilic than chitosan, which is a natural polymer [41,50].

Table 2. MC, WVDC, and mechanical properties (E, σ , and ϵ) of the films.

Film	MC (%) *	WVDC ($\times 10^{-8} \text{ cm}^2/\text{s}$) *	E (MPa) *	σ (MPa) *	ϵ (%) *
CSNP	30 ± 2^c	0.152 ± 0.020^a	51.85 ± 8.72^b	2.88 ± 0.86^b	6.95 ± 0.88^a
PVA	4 ± 1^a	23.101 ± 0.804^f	67.44 ± 12.11^c	24.68 ± 1.15^c	124.66 ± 7.75^d
30PVA/0.125OPB	7 ± 1^{bd}	7.668 ± 0.107^d	11.26 ± 0.29^a	1.32 ± 0.23^a	17.37 ± 2.81^{bc}
30PVA/0.25OPB	7 ± 1^{bd}	8.272 ± 0.268^d	10.52 ± 1.03^a	1.21 ± 0.12^a	11.97 ± 2.88^{ab}
30PVA/0.5OPB	7 ± 1^b	9.654 ± 0.242^e	17.34 ± 2.54^a	1.11 ± 0.07^a	11.50 ± 1.50^{ab}
40PVA/0.125OPB	6 ± 1^b	1.431 ± 0.319^b	8.84 ± 0.37^a	1.21 ± 0.26^a	22.37 ± 4.32^c
40PVA/0.25OPB	7 ± 2^b	2.686 ± 0.154^c	10.86 ± 1.10^a	0.81 ± 0.15^a	11.58 ± 1.83^{ab}
40PVA/0.5OPB	9 ± 2^d	3.585 ± 0.249^c	12.34 ± 0.62^a	0.72 ± 0.05^a	9.49 ± 1.04^a

* Different letters mean significant differences ($p < 0.05$) determined by Tukey's test (WVDC, E, σ , and ϵ) and LSD test for MC. MC = moisture content; WVDC = water vapor diffusion coefficient; E = Young's modulus; σ = tensile strength; ϵ = elongation at break.

For the WVDC (Table 2), the PVA film showed the highest value and CSNP showed the lowest, respectively. This is in agreement with previous research [21]. For the blends, the WVDC increased as the OPB increased. This behavior was also reported by Terzioğlu et al. [6] with the addition of orange peel to a CS/PVA matrix. This was attributed to the heterogeneity of the film due to the incorporation of the hydrophilic orange peel [20,51]. In contrast, as CS content decreased for the 40% PVA blend films (see Table 1), the WVDC decreased compared to the 30% PVA blends. This is associated with the more hydrophilic nature of CS as previously mentioned. Hydrophilic films exhibit higher water vapor diffusion than hydrophobic ones [52]. For the WVDC, statistical differences were found ($p < 0.05$).

From the mechanical properties (Table 2), Young's modulus and tensile strength were higher for PVA compared to CSNP. The elongation at break was also higher for PVA. Pure PVA has strong intermolecular hydrogen bonding due to -OH functional groups with $\sigma = 24.68$ MPa. Although NPs act as filler in the CSNP film, there is a disruption in the CS network causing uneven stress transmission with a low value of tensile strength (2.88 MPa). For the blends, strong polymer interactions in PVA are broken; although new

interactions are formed between the polymers (CS and PVA) and the OPB, improving miscibility, microdefects as pores, defects, and cracks occur (as seen in SEM micrographs in Figure 2), decreasing stress transfer and lowering tensile strength due to crack initiation at the OPB/polymer matrix interface [53,54].

On the other hand, Young's modulus and tensile strength were higher for PVA compared to CSNP. The elongation at break was also higher for PVA. For the blends, Young's modulus and tensile strength decreased compared to both PVA and CSNP. The elongation at break slightly increased compared to the CSNP film. As the OPB percentage increased, Young's modulus increased and the elongation at break decreased, showing the reinforcing effect of the filler. The hydrogen bonding interaction in the blend is given by the interaction between -OH and -NH₂ groups of CS and -OH groups of PVA, between -OH and -NH₂ groups of CS and -OH and -COOH groups of OPB, and between -OH groups of PVA and -OH and -COOH groups of OPB. At low OPB content, hydrogen bonding was favored between CS and PVA, with higher flexibility causing lower Young's modulus and higher elongation at break. As OPB content increased, stronger hydrogen bonding between components reduced chain mobility, increasing stiffness and decreasing elongation at break.

As the percentage of PVA increased for the 40% PVA blends, Young's modulus decreased and the elongation at break increased due to PVA flexibility. This behavior was also reported by other authors [6,18,55,56]. Due to interfacial interactions, OPB modifies chain stiffness and mobility, resulting in changes to Young's modulus and elongation at break. On the other hand, tensile strength is dependent on stress transfer efficiency [57]. The tensile strength values of the blends were not significantly different ($p < 0.05$). This behavior can be attributed to the low OPB concentrations (up to 0.5 wt%) and to the limited interfacial adhesion between the filler and the polymeric matrix. Two opposite phenomena occurred: the reinforcing effect (hydrogen bonding) and weakening (microdefects). Therefore, the filler acted as a discontinuous phase without generating sufficient stress transfer to modify tensile strength [58]. This was previously discussed in Section 3.2. Although tensile strength remained stable, the selection of formulations based on other functional or environmental criteria such as WVDC or biodegradability should be considered.

3.5. Degradation Test

Soil pH and conductivity monitoring were measured before and after composting. Similar values were obtained. pH values were 8.5 ± 0.3 and 8.8 ± 0.7 , before and after composting, respectively. For conductivity, values of -91.0 ± 12.9 mV and -95.7 ± 11.2 mV were found. Also, the water activity (A_w) of the substrate in which the films were composted was monitored to determine the water conditions to which the films were subjected. Results were as follows: 0.82 ± 0.01 (day 1), 0.82 ± 0.01 (day 8), 0.83 ± 0.01 (day 15), and 0.83 ± 0.01 (day 30).

Regarding the changes in the composition of microorganisms during compost for mesophilic to thermophilic bacteria, on day 0 it was 1×10^6 CFU g⁻¹ for mesophilic and 0.4×10^6 CFU g⁻¹ for thermophilic. On day 30, these values changed to 2.4×10^6 CFU g⁻¹ for mesophilic and 3.6×10^6 CFU g⁻¹ for thermophilic, with an increase in thermophilic bacteria. For fungi and yeasts, no significant changes were obtained. During the composting process, changes occur in the quantity and composition of microorganisms due to temperature changes that take place in the compost because of enzymatic degradation processes [59].

Weight loss of the films can be seen in Figure 5. After 4 weeks of soil burial, weight loss was more than 93% for the nanostructured films, being 100% for the PVA film. Therefore, biodegradation and disintegration were completed for a period of less than 180 days. Significant statistical differences were observed between weeks ($p < 0.05$).

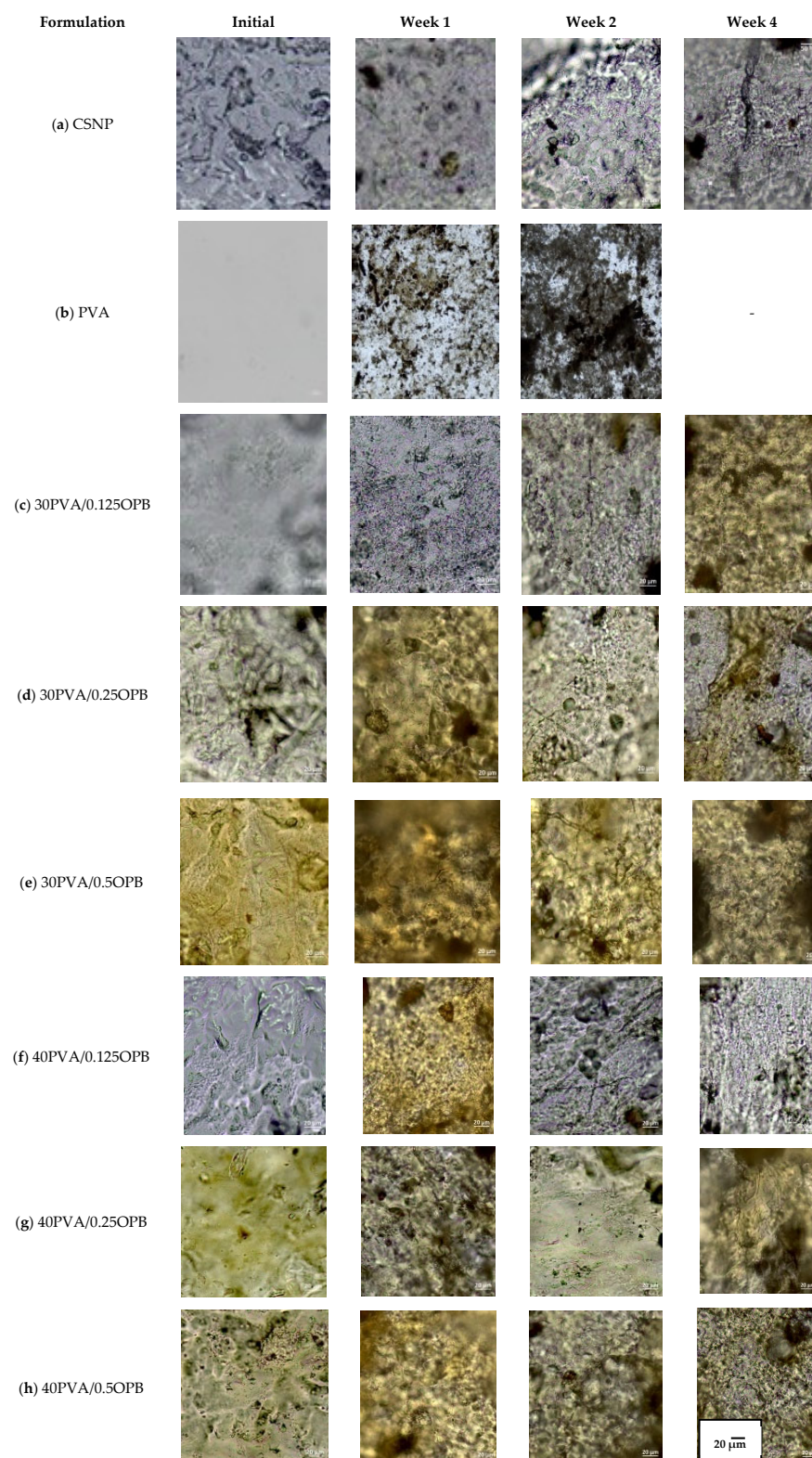


Figure 6. CLSM surface film micrographs of (a) CSNP, (b) PVA, (c) 30PVA/0.125OPB, (d) 30PVA/0.25OPB, (e) 30PVA/0.5OPB, (f) 40PVA/0.125OPB, (g) 40PVA/0.25OPB, (h) 40PVA/0.5OPB of films after 4 weeks of composting.

For the ATR-FTIR results shown in Figure 7, some changes in the main characteristic peaks were observed with degradation time. For CSNP, the decrease in the peaks at 1630 and 1020 cm^{-1} could be related to the film's degradation, caused by the breakdown of β -(1–4) glycosidic bonds (C–O–C) by microbial enzymes such as $\text{exo-}\beta$ -d-glucosaminidase

and chitosanase [66–68]. The decrease in the broad band between 3500 and 3000 cm^{-1} is also related to enzymatic activity in the soil [69]. For PVA, the decrease or disappearance of peaks (see Section 3.3) is associated with random carbon chain scission from the polymer backbone and enzyme action [70,71]. For the blends, the intensity decreased for the peaks located between 3625 and 3005 cm^{-1} , at 2919 cm^{-1} , at 1729 cm^{-1} , at 1621 cm^{-1} , at 1521 cm^{-1} , and between 1160 and 885 cm^{-1} , corresponding to the symmetric and asymmetric stretching of the -OH groups characteristic of lignin, pectin, cellulose, and polyphenolic compounds, -CH stretching, carboxylate ions (COO^-) stretching of pectin, $\text{C}=\text{C}$ stretching vibration in aromatic rings, and to $\text{C}-\text{O}-\text{H}$ vibration and out-of-plane and in-plane $\text{C}-\text{H}$ bending for aromatics, respectively. This can be attributed to the removal of lignocellulose and hemicellulose from the blends during composting due to the enzymatic action of microorganisms [17,60,72].

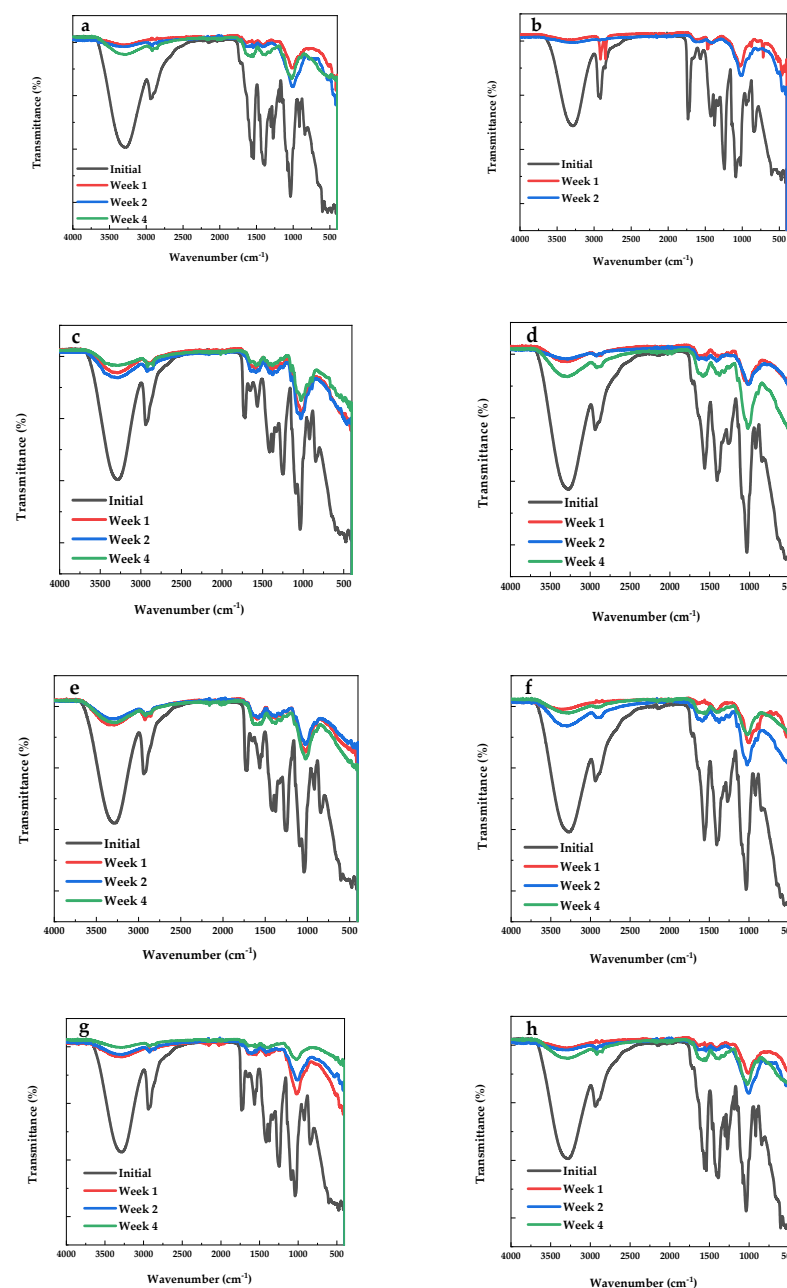


Figure 7. FTIR spectra vs. degradation time (weeks) for the different formulations: (a) CSNP, (b) PVA, (c) 30PVA/0.125OPB, (d) 30PVA/0.25OPB, (e) 30PVA/0.5OPB, (f) 40PVA/0.125OPB, (g) 40PVA/0.25OPB, (h) 40PVA/0.5OPB (see Table 1).

4. Conclusions

The incorporation of OPB into the CS/PVA matrix resulted in concentration-dependent morphological and structural modifications, as evidenced by SEM and CLSM. ATR-FTIR spectroscopy confirmed molecular interactions among chitosan, PVA, and OPB through hydrogen bonding and imine bond formation. Increasing OPB concentration enhanced WVDC and Young's modulus, while simultaneously decreasing elongation at break. The 40% PVA formulations exhibited superior flexibility compared to 30% PVA blends. Biodegradation tests showed a weight loss higher than 93% after four weeks of soil burial, while pure PVA achieved complete disintegration. The incorporation of OPB enhanced biodegradability. CLSM and ATR-FTIR analyses confirmed these results. The formulation that demonstrated the most suitable balance of mechanical, barrier, and biodegradation properties for fresh produce preservation was the 30PVA/0.25OPB film due to an intermediate value of WVDC, adequate flexibility, structural integrity, and biodegradation behavior compared to pure PVA and a higher value and CSNP. The results valorize the use of agricultural by-products through their integration into biodegradable polymer matrices, thereby contributing to circular economy strategies in sustainable packaging.

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References

1. Hameed, A.Z.; Raj, S.A.; Kandasamy, J.; Baghdadi, M.A.; Shahzad, M.A. Chitosan: A Sustainable Material for Multifarious Applications. *Polymers* **2022**, *14*, 2335. [\[CrossRef\]](#)
2. Harutyunyan, L.R.; Lasareva, E.V. Chitosan and its derivatives: A step towards green chemistry. *Biointerface Res. Appl. Chem.* **2023**, *13*, 578. [\[CrossRef\]](#)
3. Flórez, M.; Guerra-Rodríguez, E.; Cazón, P.; Vázquez, M. Chitosan for food packaging: Recent advances in active and intelligent films. *Food Hydrocoll.* **2022**, *124*, 107328. [\[CrossRef\]](#)
4. Liu, T.; Li, J.; Tang, Q.; Qiu, P.; Gou, D.; Zhao, J. Chitosan-based materials: An overview of potential applications in food packaging. *Foods* **2022**, *11*, 1490. [\[CrossRef\]](#)
5. Dirpan, A.; Ainani, A.F.; Djalal, M. A review on biopolymer-based biodegradable film for food packaging: Trends over the last decade and future research. *Polymers* **2023**, *15*, 2781. [\[CrossRef\]](#)
6. Terzioğlu, P.; Güney, F.; Parın, F.N.; Şen, I.; Tuna, S. Biowaste orange peel incorporated chitosan/polyvinyl alcohol composite films for food packaging applications. *Food Packag. Shelf Life* **2021**, *30*, 100742. [\[CrossRef\]](#)
7. Zhang, W.; Khan, A.; Ezati, P.; Priyadarshi, R.; Sani, M.A.; Rathod, N.B.; Goksen, G.; Rhim, J.W. Advances in sustainable food packaging applications of chitosan/polyvinyl alcohol blend films. *Food Chem.* **2024**, *443*, 138506. [\[CrossRef\]](#)
8. Khezerlou, A.; Tavassoli, M.; Alizadeh Sani, M.; Mohammadi, K.; Ehsani, A.; McClements, D.J. Application of nanotechnology to improve the performance of biodegradable biopolymer-based packaging materials. *Polymers* **2021**, *13*, 4399. [\[CrossRef\]](#)
9. Gutiérrez-Molina, J.; Corona-Rangel, M.L.; Ventura-Aguilar, R.I.; Barrera-Necha, L.L.; Bautista-Baños, S.; Correa-Pacheco, Z.N. Chitosan and *Byrsonima crassifolia*-based nanostructured coatings: Characterization and effect on tomato preservation during refrigerated storage. *Food Biosci.* **2021**, *42*, 101212. [\[CrossRef\]](#)

10. Correa-Pacheco, Z.N.; Corona-Rangel, M.L.; Bautista-Baños, S.; Ventura-Aguilar, R.I. Application of natural-based nanocoatings for extending the shelf life of green bell pepper fruit. *J. Food Biosci.* **2021**, *86*, 95–102. [\[CrossRef\]](#)
11. Istúriz-Zapata, M.A.; Correa-Pacheco, Z.N.; Bautista-Baños, S.; Acosta-Rodríguez, J.L.; Hernández-López, M.; Barrera-Necha, L.L. Efficacy of extracts of mango residues loaded in chitosan nanoparticles and their nanocoatings on in vitro and in vivo postharvest fungal. *J. Phytopathol.* **2022**, *170*, 661–674. [\[CrossRef\]](#)
12. Wang, Z.; Chen, X.; Guo, Z.; Feng, X.; Huang, P.; Du, M.; Zalán, Z.; Kan, J. Distribution and natural variation of free, esterified, glycosylated, and insoluble-bound phenolic compounds in brocade orange (*Citrus sinensis* L. Osbeck) peel. *Food Res. Int.* **2022**, *153*, 110958. [\[CrossRef\]](#) [\[PubMed\]](#)
13. Ayala, J.R.; Montero, G.; Campbell, H.E.; García, C.; Coronado, M.A.; León, J.A.; Sagaste, C.A.; Pérez, L.J. Extraction and characterization of orange peel essential oil from Mexico and United States of America. *J. Essent. Oil Bear. Plants* **2017**, *20*, 897–914. [\[CrossRef\]](#)
14. Galindo-Segura, L.A.; Pérez-Vázquez, A.; Ramírez-Martínez, A.; López-Romero, G.; Gómez-Merino, F.C. El Manejo del Bagazo de Naranja en la Zona Centro del Estado de Veracruz. *Terra Lat.* **2023**, *41*, 1–8. [\[CrossRef\]](#)
15. De Medina-Salas, L.; Giraldi-Díaz, M.R.; Castillo-González, E.; Morales-Mendoza, L.E. Valorization of orange peel waste using precomposting and vermicomposting processes. *Sustainability* **2020**, *12*, 7626. [\[CrossRef\]](#)
16. Yaradoddi, J.S.; Banapurmath, N.R.; Ganachari, S.V.; Soudagar, M.E.M.; Sajjan, A.M.; Kamat, S.; Mujtaba, M.A.; Shettar, A.S.; Anqi, A.E.; Safaei, M.R.; et al. Bio-based material from fruit waste of orange peel for industrial applications. *J. Mater. Res. Technol.* **2022**, *17*, 3186–3197. [\[CrossRef\]](#)
17. Yi, V.N.W.; Huey, N.C.; Peng, T.Y.; Xian, O.Z.; Hoong, S.S. Isolation of cellulose derived from orange peel and its application in biodegradable films. *Cellul. Chem. Technol.* **2021**, *55*, 311–324. [\[CrossRef\]](#)
18. Rathinavel, S.; Saravanakumar, S.S. Development and analysis of poly vinyl alcohol/orange peel powder biocomposite films. *J. Nat. Fibers* **2021**, *18*, 2045–2054. [\[CrossRef\]](#)
19. Sánchez-Orozco, R.; Timoteo-Cruz, B.; García- Sánchez, J.J.; Gomez-Espinosa, R.M.; Bernal-Martínez, L.A.; Torres-Blancas, T. Properties of eco-friendly orange peel-alginate-glycerol bioplastic films as potential food packaging applications. *J. Macromol. Sci. Part A* **2024**, *61*, 528–540. [\[CrossRef\]](#)
20. Taghavi Kevij, H.; Salami, M.; Mohammadian, M.; Khodadadi, M.; Emam-Djomeh, Z. Mechanical, physical, and bio-functional properties of biopolymer films based on gelatin as affected by enriching with orange peel powder. *Polym. Bull.* **2021**, *78*, 4387–4402. [\[CrossRef\]](#)
21. Correa-Pacheco, Z.N.; Bautista-Baños, S.; Hernández-López, M.; Tapia-Maruri, D.; Jiménez-Pérez, J.L.; Ortega-Gudiño, P.; Cruz-Miranda, L. Characterization of nanostructured chitosan-PVA films and their effects on blueberries during storage. *Future Foods* **2025**, *11*, 100571. [\[CrossRef\]](#)
22. Correa-Pacheco, Z.N.; Bautista-Baños, S.; Corona-Rangel, M.L.; Ventura-Aguilar, R.I.; Jiménez-Pérez, J.L.; Cruz-Orea, A.; Fonseca-García, A.; López-Gamboa, G.; Olvera-Cano, L.I. Morphological, optical and thermal properties of bioactive-chitosan nanostructured edible films for food packaging applications. *Food Biophys.* **2024**, *19*, 207–218. [\[CrossRef\]](#)
23. Sreekumar, S.; Goycoolea, F.; Moerschbacher, B.; Rivera-Rodriguez, G. Parameters influencing the size of chitosan-TPP nano- and microparticles. *Sci. Rep.* **2018**, *8*, 4695. [\[CrossRef\]](#) [\[PubMed\]](#)
24. Athavale, R.; Sapre, N.; Rale, V.; Tongaonkar, S.; Manna, G.; Kulkarni, A.; Shirolkar, M.M. Tuning the surface charge properties of chitosan nanoparticles. *Mat. Lett.* **2022**, *308*, 131114. [\[CrossRef\]](#)
25. Németh, Z.; Csóka, I.; Semnani Jazani, R.; Sipos, B.; Haspel, H.; Kozma, G.; Kónya, Z.; Dobó, D.G. Quality by design-driven Zeta potential optimisation study of liposomes with charge imparting membrane additives. *Pharmaceutics* **2022**, *14*, 1798. [\[CrossRef\]](#)
26. Karayianni, M.; Sentoukas, T.; Skandalis, A.; Pippa, N.; Pispas, S. Chitosan-based nanoparticles for nucleic acid delivery: Technological aspects, applications and future perspectives. *Pharmaceutics* **2023**, *15*, 1849. [\[CrossRef\]](#)
27. Zhang, X.; Wang, Q.; Liu, Z.; Zhi, L.; Jiao, B.; Hu, H.; Ma, X.; Agyei, D.; Shi, A. Plant protein-based emulsifiers: Mechanisms, techniques for emulsification enhancement and applications. *Food Hydrocoll.* **2023**, *144*, 109008. [\[CrossRef\]](#)
28. Wan, Q.; Thompson, B.C. Control of properties through hydrogen bonding interactions in conjugated polymers. *Adv. Sci.* **2024**, *11*, 2305356. [\[CrossRef\]](#)
29. El-Saadony, M.T.; Saad, A.M.; Sitohy, M.; Alkafaas, S.S.; Dladla, M.; Ghosh, S.; Mohammed, D.M.; Solima, T.N.; Ibrahim, E.H.; Fahmy, M.A.; et al. Chitosan nanoparticles: Green synthesis, biological activities, and sustainable frontiers in targeted drug delivery and cancer nanomedicine—A comprehensive review. *Mater. Today Bio* **2025**, *35*, 102358. [\[CrossRef\]](#)
30. Aycan, D.; Yayla, N.A.; Aydin, Y.A. Chitosan polyvinyl alcohol blend films for ibuprofen encapsulation: Fabrication, characterization and kinetics. *Polym. Degrad. Stab.* **2020**, *181*, 109346. [\[CrossRef\]](#)
31. Zhou, X.; Liu, L.; Li, J.; Wang, L.; Song, X. Extraction and characterization of pectin from Jerusalem artichoke residue and its application in blueberry preservation. *Coatings* **2022**, *12*, 385. [\[CrossRef\]](#)
32. Naik, P.; Pradhan, S.; Acharya, S.K.; Sahoo, P. Effect of carbonization of orange peel particulate-reinforced polymer composites: Mechanical and morphological properties. *Int. J. Surf. Eng. Interdiscip. Mater. Sci.* **2022**, *10*, 1–20. [\[CrossRef\]](#)

33. Tabatabaieian, A.; Ghasemi, A.R.; Shokrieh, M.M.; Marzbanrad, B.; Baraheni, M.; Fotouhi, M. Residual stress in engineering materials: A review. *Adv. Eng. Mater.* **2022**, *24*, 2100786. [\[CrossRef\]](#)
34. Bellon, J.; Bacoup, F.; Maris, S.; Gattin, R. PLA, PBS, and PBAT biocomposites -Part A: Matrix-filler interactions with agro-industrial waste fillers (brewer's spent grain, orange peel) and their influence on thermal mechanical and water sorption properties. *Materials* **2025**, *18*, 3867. [\[CrossRef\]](#)
35. Pervin, R.; Ghosh, P.; Basavaraj, M.G. Tailoring pore distribution in polymer films via evaporation induced phase separation. *RSC Adv.* **2019**, *9*, 15593. [\[CrossRef\]](#)
36. Botta, L.; Mistretta, M.C.; Lamattina, G.; Gargano, F.; Liguori, G. Opuntia-ficus indica fruit by-products as fillers for PLA-based biocomposites: A comparison between glochids and peel. *Polym. Compos.* **2025**, *46*, 12243–12256. [\[CrossRef\]](#)
37. Shirazi, R.; Mohammadi, T.; Koupaie, E.H.; De France, K.J. Chemically crosslinked electrospun chitosan/pol(vinyl alcohol) membranes with encapsulated zeolite for organic dye removal. *J. Water Process Eng.* **2025**, *77*, 108413. [\[CrossRef\]](#)
38. Yamala, A.; Pandit, R.; Kanparthi, R.K.; Katti, P.; Vallabhapurapu, S.; Pujala, R.K. Physically crosslinked poly (methacrylic acid-co-acrylamide)/gelatin chitosan (poly-MAGC) interpenetrating polymer network hydrogels for drug delivery and antibacterial activity. *Mater. Adv.* **2025**, 1–16. [\[CrossRef\]](#)
39. Wongsu, P.; Phatikulrungsun, P.; Prathumthong, S. FT-IR characteristics, phenolic profiles and inhibitory potential against digestive enzymes of 25 herbal infusions. *Sci. Rep.* **2022**, *12*, 6631. [\[CrossRef\]](#)
40. Fang, L.; Yang, H.; Yang, J.; Peng, M.; Hu, J. Preparation and characterization of chitosan/gelatin/PVA hydrogel for wound dressings. *Carbohydr. Polym.* **2016**, *146*, 427–434.
41. Ali, M.; Gherissi, A. Synthesis and characterization of the composite material PVA/chitosan/5% sorbitol with different ratio of chitosan. *Int. J. Mech. Mechatron. Eng.* **2017**, *17*, 15–28.
42. Abdelghany, A.M.; Menazea, A.A.; Ismail, A.M. Synthesis, characterization and antimicrobial activity of chitosan/polyvinyl alcohol blend doped with *Hibiscus sabdariffa* L. extract. *J. Mol. Struct.* **2019**, *1197*, 603–609. [\[CrossRef\]](#)
43. Olvera Bernal, R.A.; Olekhovich, R.O.; Uspenskaya, M.V. Chitosan/PVA nanofibers as potential material for the development of soft actuators. *Polymers* **2023**, *15*, 2037. [\[CrossRef\]](#) [\[PubMed\]](#)
44. Raksa, A.; Utke, R.; Ruksakulpiwat, C.; Numpaisal, P.; Ruksakulpiwat, Y. Morphological and chemical characterization of electrospun silk fibroin/polyvinyl alcohol nanofibers. *AIP Conf. Proc.* **2020**, *2279*, 080004. [\[CrossRef\]](#)
45. Kadir, M.F.Z.; Aspanut, Z.; Majid, S.R.; Arof, A.K. FTIR studies of plasticized poly(vinyl alcohol)-chitosan blend doped with NH_4NO_3 polymer electrolyte membrane. *Spectrochim. Acta A* **2011**, *78*, 1068–1074. [\[CrossRef\]](#)
46. Zeng, P.; Chen, X.; Qin, Y.; Zhang, Y.; Wang, X.; Wang, J.; Ning, Z.; Ruan, Q.; Zhang, Y. Preparation and characterization of a novel colorimetric indicator film based on gelatin/polyvinyl alcohol incorporating mulberry anthocyanin extracts for monitoring fish freshness. *Food Res. Int.* **2019**, *126*, 108604. [\[CrossRef\]](#)
47. Alfuraydi, R.T.; Alminderej, F.M.; Mohamed, N.A. Evaluation of antimicrobial and anti-biofilm formation activities of novel poly(vinyl alcohol) hydrogels reinforced with crosslinked chitosan and silver nano-particles. *Polymers* **2022**, *14*, 1619. [\[CrossRef\]](#)
48. Alfuraydi, R.T.; Al-Harby, N.F.; Alminderej, F.M.; Elmehbad, N.Y.; Mohamed, N.A. Poly (vinyl alcohol) hydrogels boosted with cross-linked chitosan and silver nanoparticles for efficient adsorption of congo red and crystal violet dyes. *Gels* **2023**, *9*, 882. [\[CrossRef\]](#)
49. Chetouani, A.; Elkolli, M.; Bounekhel, M.; Benachour, D. Chitosan/oxidized pectin/PVA blend film: Mechanical and biological properties. *Polym. Bull.* **2017**, *74*, 4297–4310. [\[CrossRef\]](#)
50. Cheng, Y.; Far, B.F.; Jahanbakhshi, M.; Bahrami, S.; Tamimi, P.; Sedaghatf, M.; Ghazizadehagh, E. Exploring the potential of a polyvinyl alcohol/chitosan-based nanofibrous matrix for erythromycin delivery: Fabrication, in vitro and in vivo evaluation. *RSC Adv.* **2023**, *13*, 18450. [\[CrossRef\]](#)
51. Machado, M.I.R.; Ângelo, P.; Gonçalves, Â.; Monção, R.M.; de Souza, R.R.M.; Machado, A.R. Characterization of biodegradable films applicable to agriculture with structural reinforcement. *Eng. Proc.* **2024**, *67*, 38.
52. Wang, F.; Han, S.; Zhang, Y.; Gao, L.; Li, X.; Zhao, L.; Ye, H.; Li, H.; Xin, Q.; Zhang, Y. Constructing rapid water vapor transport channels within mixed matrix membranes based on two-dimensional mesoporous nanosheets. *Commun. Chem.* **2022**, *5*, 65. [\[CrossRef\]](#) [\[PubMed\]](#)
53. Awaja, F.; Zhang, S.; Tripathi, M.; Nikiforov, A.; Pugno, N. Cracks, microcracks and fracture in polymer structures: Formation, detection, automatic repair. *Prog. Mater. Sci.* **2016**, *83*, 536–573. [\[CrossRef\]](#)
54. Olonisakin, K.; Mohanty, A.K.; Thimmanagari, M.; Misra, M. Recent advances in biodegradable polymer blends and their biocomposites: A comprehensive review. *Green Chem.* **2025**, *27*, 11656–11704. [\[CrossRef\]](#)
55. Koutoulis, A.S.; Giannakas, A.E.; Lazaridis, D.G.; Kitsios, A.P.; Karabagias, V.K.; Giannakas, A.E.; Ladavos, A.; Karabagias, I.K. Preparation and characterization of PLA-based films fabricated with different citrus species peel powder. *Coatings* **2024**, *14*, 1311. [\[CrossRef\]](#)

56. Tone, A.M.; Herranz Solana, N.; Khan, M.R.; Borriello, A.; Torrieri, E.; Sánchez Reig, C.; Monedero Prieto, F.M. Study on the properties of PLA and PP-based films for food applications incorporating orange peel extract from agricultural by-products. *Polymers* **2024**, *16*, 1245. [[CrossRef](#)]
57. Khieng, T.K.; Debnath, S.; Ting Chaw Liang, E.; Anwar, M.; Pramanik, A.; Basak, A.K. A Review on Mechanical Properties of Natural Fibre Reinforced Polymer Composites under Various Strain Rates. *J. Compos. Sci.* **2021**, *5*, 130. [[CrossRef](#)]
58. Sathiya Narayanan, N.; Sai Venkat Mohan, D.; Abhinay, J.; Dinesh, T.; Satya Sai Surya Teja, V.; Praneeth, R. Effects on microhardness, tensile strength, deflection, and drop weight impact resistance with the addition of hybrid filler materials for enhancing GFRP composites. *Sci. Rep.* **2024**, *14*, 27524. [[CrossRef](#)]
59. Moreno, J.; López-González, J.A.; Arcos-Nievas, M.A.; Suárez-Estrella, F.; Jurado, M.M.; Estrella-González, M.J.; López, M.J. Revisiting the succession of microbial populations throughout composting: A matter of thermotolerance. *Sci. Total Environ.* **2021**, *773*, 145587. [[CrossRef](#)]
60. Ayala, J.R.; Montero, G.; Coronado, M.A.; García, C.; Curiel-Alvarez, M.A.; León, J.A.; Sagaste, C.A.; Montes, D.G. Characterization of orange peel waste and valorization to obtain reducing sugars. *Molecules* **2021**, *26*, 1348. [[CrossRef](#)]
61. Sambudi, N.S.; Lin, W.Y.; Harun, N.Y.; Mutiari, D. Modification of poly(lactic acid) with orange peel powder as biodegradable composite. *Polymers* **2022**, *14*, 4126. [[CrossRef](#)] [[PubMed](#)]
62. Das, A.; Ringu, T.; Ghosh, S.; Pramanik, N. in preparation, physicochemical characterization, and bioengineering applications of biopolymers. *Polym. Bull.* **2023**, *80*, 7247–7312. [[CrossRef](#)] [[PubMed](#)]
63. Elgharbawy, A.S.; El Demerdash, A.M.; Sadik, W.A.; Kasaby, M.A. Enhancing the biodegradability, water solubility, and thermal properties of polyvinyl alcohol through natural polymer blending: An approach toward sustainable polymer applications. *Polymers* **2024**, *16*, 2141. [[CrossRef](#)] [[PubMed](#)]
64. Liu, Y.; Wang, S.; Lan, W. Fabrication of antibacterial chitosan-PVA blended film using electrospray technique for food packaging applications. *Int. J. Biol. Macromol.* **2018**, *107*, 848–854. [[CrossRef](#)]
65. Wardhono, E.Y.; Pinem, M.P.; Susilo, S.; Siom, B.J.; Sudrajad, A.; Pramono, A.; Meliana, Y.; Guénin, E. Modification of physio-mechanical properties of chitosan-based films via physical treatment approach. *Polymers* **2022**, *14*, 5216. [[CrossRef](#)]
66. Altun, E.; Çelik, E.; Ersan, H.Y. Tailoring the Microbial Community for Improving the Biodegradation of Chitosan Films in Composting Environment. *J. Polym. Environ.* **2020**, *28*, 1548–1559. [[CrossRef](#)]
67. Mahmud, M.; Talip, N.; Yacob, N.; Idris, S. Application interval and concentration effect of gamma degraded chitosan on mulberry plant. *Food Res.* **2023**, *7*, 94–100. [[CrossRef](#)]
68. Westlake, J.R.; Chaloner, E.; Laabei, M.; Sgouridis, F.; Burrows, A.D.; Xie, M. Degradation investigation and active packaging performance of cross-linked chitosan film containing gallic acid. *RSC Sustain.* **2025**, *3*, 2680–2695. [[CrossRef](#)]
69. Oberlintner, A.; Bajić, M.; Kalčíková, G.; Likozar, B.; Novak, U. Biodegradability study of active chitosan biopolymer films enriched with Quercus polyphenol extract in different soil types. *Environ. Technol. Innov.* **2021**, *21*, 101318. [[CrossRef](#)]
70. Pantelic, B.; Ponjavic, M.; Jankovic, V.; Aleksic, I.; Stevanovic, S.; Murray, J.; Fournet, M.B.; Nikodinovic-Runic, J. Upcycling biodegradable PVA/starch film to a bacterial biopigment and biopolymer. *Polymers* **2021**, *13*, 3692. [[CrossRef](#)]
71. Chin, K.; Sam, S.T.; Ong, H.L.; Wong, Y.S.; Tan, W.K. Biodegradation improvement of bioinspired crosslinked and noncrosslinked polyvinyl alcohol nanocomposites with cellulose nanocrystals extracted from rice straw through natural soil burial exposure. *Polym. Compos.* **2022**, *43*, 6955–6965. [[CrossRef](#)]
72. Gao, Q.; Guo, L.; Li, S.; Wu, W.; Ding, J.; Xu, H.; Luo, C.; Li, J.; Li, D.; Liu, Z. Biodegradation mechanism of cellulose, hemicellulose, and lignin in bacteria-dominant aerobic composting from agricultural biomass waste: A review. *Carbohydr. Polym. Tech. Appl.* **2025**, *11*, 100879.

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