

Effect of tannic acid concentration on structural, mechanical, and antibacterial properties of polyvinyl alcohol/chitosan/pomegranate peel-based biocomposite films

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Tannic acid is a plant-derived natural crosslinker and active agent, generally improving the properties of biodegradable films. This study investigated the effect of tannic acid (TA) incorporation on the mechanical, structural, and antibacterial properties of biocomposite films based on polyvinyl alcohol (PVA), chitosan (CS), and pomegranate peel powder (PPP). Tannic acid was added to the film-forming solution at concentrations ranging from 6% to 10% (w/w), based on the total polymer weight. The mechanical performance of the films was evaluated in terms of elastic modulus, tensile strength, and elongation at break. The addition of tannic acid resulted in improved mechanical performance, as evidenced by higher elastic modulus and tensile strength values relative to those of the control film. The highest elastic modulus and tensile strength values were obtained for the films containing 10% (w/w) tannic acid. FT-IR results revealed the formation of hydrogen bonding interactions between the functional groups of tannic acid and polymers. The antibacterial activity of the control film and the film containing the highest concentration of tannic acid was evaluated against *Escherichia coli* and *Staphylococcus aureus* by the modified ISO 20645 disc diffusion method. The film samples did not exhibit any antibacterial activity.

Keywords: Polyvinyl alcohol, Chitosan, Tannic acid, Pomegranate peel powder

INTRODUCTION

Packaging is a method or material that plays a critical role in protecting products from environmental effects, extending the shelf life of food products, and enabling safe global distribution [1]. Globally, approximately half of the packaging industry consists of conventional petroleum-based plastics due to their low cost, durability, and ease of processability. Nevertheless, these conventional plastics have led to significant environmental and economic challenges such as long degradation times, micro-plastic pollution, and their high carbon footprint in production processes [2–5]. Similar to the situation with petroleum-based plastics, agricultural food waste and agro-industrial by-products also constitute a significant global problem affecting national economies, world resources, and public health [6–8]. A promising solution involves recycling/reusing/upcycling food waste and by-products to develop sustainable, biodegradable packaging, which directly aligns with circular economy strategies [2]. In this context, there is a growing demand for eco-friendly, sustainable, and functional bio-based packaging materials developed using biopolymers, biodegrad-

able polymers, and biocomposites. PVA is one of the most preferred and commonly used biodegradable polymers in biocomposite films due to its unique properties such as high chemical resistance, non-toxicity, and biocompatibility [9]. Another biodegradable and bio-based polymer commonly used, and also preferred in this study, is chitosan. Chitosan is a linear polysaccharide derived from chitin with exceptional properties such as biodegradability, biocompatibility, and antibacterial activity [9]. However, food packaging films based on a single biopolymer component may have various disadvantages that limit their practical applications and functional properties in the food packaging industry. Polymer blending is the preferred first choice and a promising strategy for the development of biodegradable food packaging films. Blending different polymers makes it possible to create films with unique properties while preserving the distinctive properties of each polymer [10, 11]. In recent years, especially in the food packaging field, biocomposite film structures have begun to be developed with various organic or inorganic additives at the micro- or nano-level, incorporated into polymer blends to improve

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several properties (thermal, barrier, mechanical, antibacterial, etc.) of the films.

Biocomposites are materials composed of at least two components, referred to as the polymer matrix and the reinforcement material, at least one of which is bio-based [12]. With the above-mentioned environmental concerns, the use of food waste (fruit peels, ground seeds, etc.) and agricultural by-products (stems, stalks, etc.) as reinforcement materials in synthetic or bio-based polymer matrices is gaining attention [13, 14].

Pomegranate peel powder, a by-product of the pomegranate fruit, has gained significant attention as an organic reinforcement material in the food packaging, cosmetic, and medical fields, owing to its valuable antioxidant, antibacterial, anti-inflammatory, and anticancer properties [15]. Pomegranate peel powder contains phenolic compounds and flavonoids such as apigenin, caffeic acid, chlorogenic acid, ellagic acid, gallic acid, quercetin, and punicalagin [17]. Active packaging films containing pomegranate peel powder exhibit excellent antimicrobial and antioxidant activity, which makes them an attractive option for extending the shelf life of food products.

Tannic acid is a natural green crosslinker used in polymeric films. Tannic acid is a plant-derived polyphenolic compound, consisting of polymerized flavonoids with varying degrees of polymerization, and containing catechol and pyrogallol aromatic rings [18]. As one of the most abundant naturally occurring compounds, it offers economic advantages and exhibits excellent biocompatibility along with antioxidant, anti-inflammatory, and antimicrobial properties [19]. Due to its rich phenolic hydroxyl content, tannic acid can readily associate with biological macromolecules through various intermolecular interactions. As a crosslinker, it has a tendency to form more stable structures [20]. In the literature, food packaging films/coatings based on biopolymers cross-linked with tannic acid have shown good effects in preserving fresh foods, as tannic acid improves the mechanical, barrier, and thermal properties of the films [21, 22].

The aim of this study is to develop and characterize PVA/CS/PPP composite films incorporated with varying concentrations of tannic acid (0, 6, 8, and 10% w/w) for food packaging applications. The physicochemical, mechanical, and antibacterial properties of the films were evaluated through Fourier-transform infrared spectroscopy (FT-IR), scanning electron microscopy (SEM), hydrophilicity measurements, mechanical testing, and antibacterial assays against *Escherichia coli* and

Staphylococcus aureus.

EXPERIMENTAL

Materials

Chitosan (CS) with a deacetylation degree of at least 75% was obtained from Sigma-Aldrich (Darmstadt, Germany). Polyvinyl alcohol (PVA, 87.8% purity) was supplied by Zag Kimya (Istanbul, Türkiye). Pomegranate peel powder (PPP) was procured from Naturşah (Izmir, Türkiye). Tannic acid (TA) was purchased from Sentez Lab. Company (Istanbul, Türkiye), while glycerol, used as a plasticizing agent, was acquired from Merck (Darmstadt, Germany).

PVA/CS/PPP and PVA/CS/PPP/TA biocomposite film production

Figure 1 illustrates the general fabrication workflow of the biocomposite film production. Initially, a 5% (w/v) PVA solution was prepared by dissolving polymer granules in distilled water at 90 °C under continuous stirring at 400 rpm for 3 h. In parallel, a 1% (w/v) chitosan (CS) solution was obtained using a 1% (v/v) aqueous acetic acid solvent at 50 °C. These two polymeric solutions were subsequently blended and stirred at 50 °C for 1 h. To prepare the control film, pomegranate peel powder (PPP) was added into the neat PVA/CS blend at 3% (w/w, based on total polymer weight) concentration. Tannic acid was used in liquid form; therefore, a 5% (w/v) tannic acid stock solution was prepared with distilled water. After stirring the PPP-loaded polymer solution at 250 rpm and 25 °C for 24 h, tannic acid was added separately at concentrations of 6, 8, and 10% (w/w). The tannic acid-containing polymer solutions were stirred again at 40 °C for 2 h. In the next step, glycerol (20% w/w) was incorporated as a plasticizer, followed by an additional 30 min of stirring at room temperature. Finally, the prepared solutions were poured into circular glass Petri dishes with a diameter of 11.5 cm and dried in an oven at 50 °C for 24 h and 35 °C for 18 h, respectively [23].

Fourier transform infrared spectroscopy (FT-IR)

The biocomposite films (PCPT-0 and PCPT-10) were analyzed by FT-IR spectroscopy (Thermo Nicolet iS50 FT-IR, USA). The measurements were taken between 500–4000 cm⁻¹ wavenumbers at 4 cm⁻¹ resolution in transmission mode [13, 23].

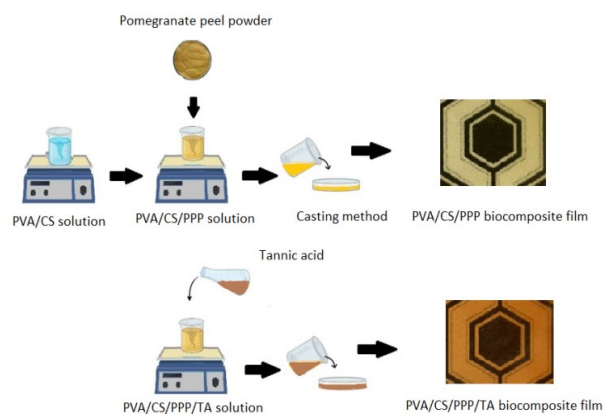


Figure 1. Schematic illustration of PPP- and TA-loaded biocomposite film preparation.

The resulting film samples were designated as PCPT-0, PCPT-6, PCPT-8, and PCPT-10, respectively, as shown in Table 1.

Table 1. Biocomposite film compositions

Sample code	PVA % (w/v)	CS % (w/v)	PPP % (w/w)	TA % (w/w)	Glycerol % (w/w)
PCPT-0	5	1	3	0	20
PCPT-6	5	1	3	6	20
PCPT-8	5	1	3	8	20
PCPT-10	5	1	3	10	20

Surface morphology analysis (SEM)

The surface morphology of the biocomposite films (PCPT-0 and PCPT-10) was analyzed on a Gemini 300 scanning electron microscope (SEM, Carl Zeiss, Germany) operating with a 5 kV acceleration voltage. The film samples were coated with gold before scanning to eliminate charge effects [13, 23].

Mechanical properties

The tensile properties of the biocomposite films were presented in terms of tensile strength, elongation at break, and Young's modulus. The tests were performed according to the ASTM D882 standard with some modifications using a 1 kN load cell and a 15 mm/min crosshead speed. A one-way ANOVA test was performed in the statistical analyses [13, 23].

Water contact angle measurements

Water contact angle measurements were conducted to determine the hydrophobic or hydrophilic character of the biocomposite films. An automatic dispensing unit coupled to an optical tensiometer (Biolin Scientific, Gothenburg, Sweden) was employed for the measurements. The measurements were taken from three points on one

film, and the results were given as average data with standard deviations [13, 23].

Antibacterial activity

The antibacterial activity of the biocomposite films (PCPT-0 and PCPT-10) was evaluated against ATCC 25922 *E. coli* and ATCC 6538 *S. aureus* bacteria using the modified ISO 20645 agar diffusion method. Bacterial suspensions prepared from test microorganisms were adjusted to a final concentration of 10^5 CFU/mL. A 1 mL aliquot of this prepared inoculum was homogeneously spread onto the surface of Mueller Hinton Agar (BD, Germany) using a sterile swab. Then, the film samples were cut into 2×2 cm dimensions and placed onto the agar surface. After the plates were gently pressed to ensure full contact of the samples with the surface, they were left for incubation at 37°C for 18–24 h. At the end of the incubation period, the clear zones formed around the film samples were measured in millimeters by two independent researchers, and the antibacterial activity was evaluated.

RESULTS AND DISCUSSION

FT-IR analysis of the biocomposite films

The FT-IR spectra of tannic acid, control film (PCPT-0), and the film with the highest tannic acid concentration (PCPT-10) are given in Figure 2. The band shift and intensity alterations around 3000–3500 cm^{-1} (O-H stretching) could be attributed to the increase in hydrogen bonding between the polymer matrix and the tannic acid functional groups. Peaks at 1605 and 1519 cm^{-1} in the spectrum of TA are classic indicators of the C=C stretching peak and aromatic ring vibrations [24, 25]. The FT-IR spectrum of the film loaded with 10% (w/w) tannic acid showed a new peak around 1622–1615 cm^{-1} compared to the control film. This newly formed peak could be attributed to the stretching vibrations of the aromatic C=C bonds originating from the polyphenolic structure of tannic acid or amide I/C=O stretching of chitosan. Moreover, this band could be the result of the shifted carbonyl (C=O) stretching vibrations, involved in intermolecular hydrogen bonding between the polymer matrix and phenolic hydroxyl groups (-OH) of tannic acid [24, 26].

SEM analysis of the biocomposite films

The micrographs of the surface views of PCPT-10 and PCPT-0 biocomposite films are shown in Figure 3. As can be seen from the SEM images, the control film has a rougher and slightly heterogeneous surface; however, PCPT-10 exhibits

a smooth, uniform, and continuous surface morphology without any visible phase separation. The reason for this could be the good compatibility and homogeneous dispersion of tannic acid within the PVA/CS/PPP polymer matrix. This morphological result is supported by literature findings and attributed to the abundance of (-OH) groups [24, 25].

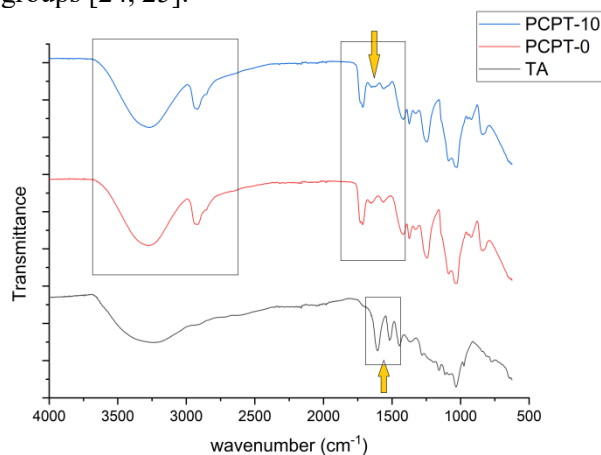


Figure 2. FT-IR spectra of tannic acid powder and biocomposite films

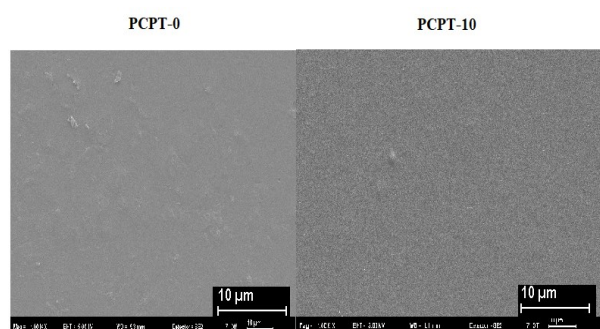


Figure 3. SEM images of the surface morphology of the biocomposite films

Mechanical properties of biocomposite films

Mechanical test results are presented in Table 2. It can be seen that tannic acid addition significantly enhanced the tensile strength and Young's modulus, thus the stiffness, of the PVA/CS/PPP biocomposite films, whereas the elongation at break values remained nearly unchanged ($p > 0.05$). The highest values of tensile strength and Young's modulus were obtained for the PCPT-10 sample, owing to the highest tannic acid concentration. This enhancement could be due to the improved intermolecular interactions such as hydrogen bonding established between the hydroxyl groups of TA and the polymer matrix, PVA/CS/PPP [26]. These interactions could lead to the development of a more compact and rigid network within the polymeric structure [24]. Initially, the elongation at break value of the 6% (w/w) tannic acid-containing

film increased compared to the control film. Then, the elongation at break values remained nearly unchanged; the control film, PCPT-0, had a value of 290.18%, and the film with the highest tannic acid concentration, PCPT-10, had 292.56%. Tensile strength and Young's modulus values of PCPT-10 reached the highest levels ($p < 0.05$) among the films. It can be said that tannic acid addition improves the mechanical strength and stiffness of the biocomposite films. In the literature, tensile strength and Young's modulus values increased with an increase in tannic acid concentration [27, 28]. However, at low tannic acid concentrations, the elongation at break value stayed almost the same compared with the control film [27].

Table 2. Mechanical test results of biocomposite films

Sample code	TA content (%)	Young's modulus (MPa)	Tensile strength (MPa)	Elongation at break (%)
PCPT-0	0	25.01 ^c ± 1.88	20.23 ^b ± 2.96	290.18 ^a ± 65.65
PCPT-6	6	34.11 ^b ± 3.11	22.87 ^{ab} ± 3.06	298.25 ^a ± 42.11
PCPT-8	8	31.44 ^{bc} ± 3.16	21.49 ^{ab} ± 2.06	290.01 ^a ± 40.81
PCPT-10	10	48.90 ^a ± 6.07	25.33 ^a ± 1.28	292.56 ^a ± 33.68

Water contact angle measurement of biocomposite films

Water contact angle is a parameter that defines the hydrophilic ($\theta < 90^\circ$) or hydrophobic ($\theta > 90^\circ$) character of a material. Contact angle values and droplet images are shown in Figure 4. It can be seen that the water contact angle values decreased from 83.1° in PCPT-0 to 75.75° in PCPT-10 with tannic acid addition. This reduction indicates an increase in the hydrophilicity of the film surfaces, which can be directly related to the hydrophilic hydroxyl (-OH) groups introduced by the tannic acid molecules [25, 28].

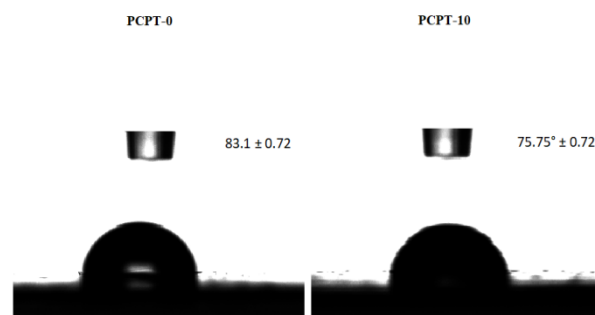


Figure 4. Water contact angle of biocomposite films

Antibacterial activity of biocomposite films

The antibacterial activity of the biocomposite films, PCPT-0 and PCPT-10, was evaluated against *E. coli* and *S. aureus* using the modified disc diffusion method on Mueller-Hinton Agar. Both the control film (PCPT-0) and the tannic acid-incorporated film (PCPT-10) swelled in the agar, and PCPT-10 migrated through the agar surface. Despite this physical migration into the agar, no zones of inhibition were observed for any of the film samples. The highly hydrophilic character of tannic acid (contact angle of 75.75°) could trigger rapid water uptake and macroscopic dissolution on the agar. Even in the dissolved state, the active phenolic hydroxyl groups of TA might be locked within the polymer-tannic acid macromolecular structure, or they might migrate in insufficient quantity to inhibit bacterial growth [29, 30]. Consequently, the active sites of tannic acid and bioactive molecules might never be released freely to interact with the bacterial cell membranes to inhibit their activity [29, 31]. In the literature, there are limited studies showing that bioactive compounds do not inhibit bacterial growth. These studies attribute this situation to the release profile of bioactive compounds and dose dependency [24, 32].

CONCLUSION

In this study, eco-friendly biocomposite films based on PVA/CS polymer blends enriched with pomegranate peel powder and different concentrations of tannic acid were successfully produced by the solution casting method for sustainable food packaging applications. Structural, mechanical, and antibacterial analyses were performed. FT-IR and SEM analyses confirmed a homogeneous and continuous film structure with good molecular compatibility between the polymer matrix and the fillers. The tannic acid incorporation improved the mechanical properties of the films, significantly increasing Young's modulus (up to 48.90 MPa) and tensile strength; however, no significant difference was observed in the elongation at break values compared to the control film. Although the tannic acid addition increased the hydrophilic character (water contact angle 75.75°) of the films due to the presence of phenolic hydroxyl groups and led to film dissolution during the modified disc diffusion tests, no antibacterial inhibition zones were observed against *E. coli* and *S. aureus*. Future studies should focus on the evaluation of antioxidant activity, barrier properties, and food shelf-life tests of the developed biocomposite films to characterize their

preservation potential. Moreover, antibacterial tests should be performed according to the ASTM E2149 Dynamic Contact Method as a complementary approach. The multifunctional biocomposite films could be sustainable alternatives to conventional synthetic packaging, with the potential to improve food preservation and reduce environmental impact; moreover, tannic acid could be a good alternative as a low-cost natural crosslinker for the polymer systems.

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