GREEN CHITOSAN/PEG/PVA PACKAGING FILMS REINFORCED WITH FRUIT PEEL ADDITIVES

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Keywords

Green packaging
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Mechanical Durability

Abstract

Packaging refers to the process applied to protect materials against environmental factors such as oxygen, moisture, light, and microorganisms, thereby preserving product quality and extending shelf life. Petroleum-based plastics, which are commonly used in packaging applications, are not biodegradable and can persist in the environment for long periods. This has led to an increasing demand for environmentally friendly and sustainable packaging materials. However, many biodegradable packaging materials exhibit limited mechanical strength, restricting their practical applicability in protective packaging systems. In this study, bio-based biocomposite films were prepared and characterized using chitosan, polyethylene glycol (PEG), and polyvinyl alcohol (PVA) biopolymers, reinforced with locally sourced hawthorn (Crataegus spp.) and pomegranate (Punica granatum L.), with the aim of developing potential biodegradable packaging films with both sufficient mechanical strength and antibacterial functionality. The results demonstrated that the biocomposite films exhibited resistance to solvents with different characteristics. Optical microscopy and SEM analyses confirmed the homogeneous dispersion of the reinforcing materials within the polymer matrix. Compared to similar materials reported in the literature, the developed biocomposite films displayed superior mechanical performance. Soil burial tests further verified that the biocomposite films exhibited significantly higher biodegradability than conventional plastics. Among the developed formulations, the 3BP film containing pomegranate peel exhibited promising characteristics for sustainable packaging applications due to its strong mechanical and antibacterial performance.

MEYVE KABUĞU KATKILARIYLA GÜÇLENDİRİLMİŞ YEŞİL KİTOSAN/PEG/PVA AMBALAJ FİLMLERİ

Anahtar Kelimeler

Yeşil ambalaj
Biyokompozit filmler
Antibakteriyel aktivite
Doğal katkılar
Mekanik dayanım

Öz

Malzemelerin oksijen, nem, ışık ve mikroorganizmalar gibi çevresel etkenlere karşı korunması; ürün kalitesinin muhafaza edilmesi ve raf ömrünün uzatılması amacıyla gerçekleştirilen işleme ambalajlama denir. Günümüzde ambalajlamada yaygın olarak kullanılan petrol kökenli plastikler biyobozunur değildir ve doğada uzun süre bozunmadan kalabilmektedir. Bu durum, çevre dostu ve sürdürülebilir ambalaj malzemelerine olan gereksinimi ortaya koymaktadır. Bununla birlikte, birçok biyobozunur ambalaj malzemesi düşük mekanik dayanım göstermekte olup, bu durum bu malzemelerin koruyucu ambalaj uygulamalarındaki kullanımını sınırlandırmaktadır. Bu çalışmada, kitosan, polietilen glikol (PEG) ve polivinil alkol (PVA) biyopolimerleri ile yerli alıç (Crataegus spp.) ve nar (Punica granatum L.) kullanılarak, yeterli mekanik dayanım ve antibakteriyel özellik gösterebilen potansiyel biyoambalajların geliştirilmesi amacıyla biyolojik temelli biyokompozit filmler hazırlanmış ve karakterize edilmiştir. Yapılan analizler, biyokompozit filmlerin farklı özelliklere sahip çözücülere karşı dayanıklılık gösterdiğini ortaya koymuştur. Optik mikroskop ve SEM görüntüleri, takviye malzemelerinin polimer matris içerisinde homojen şekilde dağıldığını doğrulamıştır. Literatürde raporlanan benzer malzemelerle karşılaştırıldığında, biyokompozitlerin daha üstün mekanik performans sergilediği belirlenmiştir. Toprağa gömme testleri sonucunda, geliştirilen biyokompozitlerin ticari plastiklere kıyasla yüksek biyobozunurluk gösterdiği tespit edilmiştir. Geliştirilen formülasyonlar arasında, nar kabuğu içeren 3BP filmi, hem mekanik hem de antibakteriyel açıdan başarılı performansı sayesinde sürdürülebilir ambalaj uygulamaları için umut verici özellikler sergilemiştir.

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1. Introduction

Plastic materials are engineering substances produced by formulating high molecular weight polymer matrices with various additives, and they function as the continuous phase in biocomposite systems (Andrady and Neal, 2009). In biocomposite applications, plastics provide a matrix environment that mechanically supports natural fibers or bio-based fillers and thus plays a decisive role in determining the overall performance and functional properties of the material (Faruk, Bledzki, Fink and Sain, 2012). Due to their advantageous characteristics such as low density, high durability, cost effectiveness, and ease of processing, plastic materials are widely utilized in numerous sectors including packaging, automotive, construction, electrical and electronic industries, and medical applications.

Composite materials are generally formed by the combination of a matrix and a reinforcing phase. The matrix phase maintains structural integrity and enables efficient load transfer, while the reinforcing phase governs the strength and stiffness characteristics of the composite. Owing to this synergistic structure, composites can exhibit mechanical and physical performances that surpass those of their individual constituents. Composite materials are commonly classified according to the geometry of the reinforcing material as fiber, particulate, and laminated composites, and according to the matrix type as polymer, metal, and ceramic composites. Among these, polymer-based fiber reinforced composites have found widespread applications due to their high specific strength and favorable processability (Hsissou et al., 2021; Sharma, Bhandari, Sharma, Dhakad and Pinca-Bretotean, 2022; Ulcay, Akyol and Gemci, 2002). Biocomposites represent the environmentally friendly class of composite materials and are defined as composites in which a natural component is employed as the reinforcing material. (Mohanty, Misra and Drzal, 2002; Pickering, Efendy and Le, 2016).

Packaging is an engineering system of critical importance across numerous sectors, including food, pharmaceutical, and electronics industries, as it ensures the protection, storage, transportation, and safe commercialization of products from production to consumption. Packaging materials play a key role in preserving product quality and extending shelf life by providing effective barriers against oxygen, moisture, light, and microorganisms (Reichert et al., 2020). The packaging industry represents the largest application area for plastics. In particular, food packaging systems are essential for mechanical protection of products and for prolonging their shelf life (Marsh and Bugusu, 2007). The widespread use of plastics in packaging applications can be attributed to their low density and resulting lightweight nature, cost efficient processing, and favorable strength to weight ratios (Karakuş, Balballı, Ara and Ayhan, 2021; Ragaert, Delva and Van Geem, 2017; Thompson, Moore, Vom Saal and Swan, 2009). Despite these advantages, the majority of petroleum-based plastics are not biodegradable (Hopewell, Dvorak and Kosior, 2009). Moreover, conventional plastics pose significant environmental concerns due to their production conditions and their long persistence in the environment without degradation. When the challenges associated with plastic waste recycling are also considered, the adoption of bioplastics and biocomposites derived from renewable resources, which play an effective role in mitigating environmental pollution, has become increasingly attractive (Karakuş et al., 2021; Huang, Dong, Che, Li and Tang, 2025). In this context, biocomposite packaging materials reinforced with natural fibers, biopolymers, and agricultural residues are regarded as environmentally friendly alternatives that can provide suitable mechanical performance and barrier properties (Prasad et al., 2025).

Numerous studies demonstrate that the incorporation of agricultural byproducts into biopolymer matrices, particularly polylactic acid (PLA) based systems, enables the production of biocomposite films with enhanced mechanical, antioxidant, and barrier properties suitable for food packaging applications (Ersus, Melikoğlu, & Cesur, 2020; Petaloti and Achilias, 2024). The increasing environmental concerns associated with petroleum-based packaging materials, particularly their non-biodegradable nature and long-term ecological persistence, have accelerated the development of sustainable biopolymer-based alternatives for food and consumer product packaging applications (Cruz et al., 2022; Gupta, Biswas and Roy, 2022). Thus, biodegradable packaging materials containing natural additives extend the shelf life of food products and serve as environmentally viable alternatives to conventional plastics. In addition, PLA based biocomposites exhibit significant potential for sustainable packaging design due to their favorable transparency, processability, and gas barrier performance (Auras, Harte and Selke, 2004; Siracusa, Rocculi, Romani and Dalla Rosa, 2008; Sorrentino,

Gorrasi and Vittoria, 2007). Furthermore, the incorporation of natural additives derived from fruit waste into biopolymer matrices emerges as an effective approach for improving the functional properties of packaging materials. Pomegranate peel, owing to its high antioxidant content, enhances UV protective and antimicrobial properties in biocomposite films (Baniasadi et al., 2025; Gil, Tomás-Barberán, Hess-Pierce, Holcroft and Kader, 2000; Singh, Kim and Lee, 2022). Similarly, hawthorn exhibits antioxidant activity depending on the extraction method and contributes to the overall performance of composite materials (Shortle et al., 2014). Owing to these attributes, pomegranate peel and hawthorn can serve as effective biodegradable functional additives in sustainable packaging technologies. Recent studies have emphasized that natural bioactive additives not only improve antioxidant and antimicrobial performance but may also affect the physical and mechanical characteristics of packaging films depending on their chemical composition and compatibility with the polymer matrix (Nasution, Harahap, Julianti, Safitri and Jaafar, 2023; Soleimanzadeh et al., 2024). Although many studies primarily focus on the biological activity of biodegradable packaging systems, maintaining sufficient mechanical strength and flexibility remains a critical requirement for practical packaging applications, particularly in protective and refillable packaging designs (Gupta et al., 2022; Pandey, Sharma and Gundabala, 2022).

In this study, the development of refillable packaging materials used in the cosmetic industry is targeted in order to reduce the environmental impacts of polymer composite materials and to enhance their applicability in sustainable engineering practices. For this purpose, biocomposite films are prepared using three different biopolymers, namely chitosan, polyethylene glycol (PEG), and polyvinyl alcohol (PVA), together with two natural supporting materials, locally sourced hawthorn (Crataegus spp.) and pomegranate (Punica granatum L.). Hawthorn and pomegranate peel were introduced into the biopolymer matrix as natural additives that may impart antibacterial functionality to the resulting materials. Unlike many previous studies mainly centered on antioxidant or antimicrobial performance, the present study comparatively evaluates the effect of two different plant-derived additives on both the biological and mechanical properties of biocomposite films intended for sustainable packaging applications (Nasution et al., 2023; Soleimanzadeh et al., 2024). The films are characterized in terms of solvent resistance, moisture retention, swelling behavior, surface contact angle, mechanical strength, biodegradability, light transmittance, and toxicity. The results of these analyses indicate that the biocomposite films represent a promising candidate for packaging applications.

2. Materials & Methods

2.1. Materials

This study was conducted in accordance with research and publication ethics. Polyvinyl alcohol (PVA) (Acros Organics), chitosan (Sigma-Aldrich), polyethylene glycol (PEG 400) (Kollisov), corn starch, and glycerol were used in the preparation of the biocomposite films. The corn starch was food-grade and obtained from local markets, while glycerol was purchased from a local pharmacy. Acetic acid (Merck) and deionized water were employed as solvents. Fresh fruits (hawthorn and pomegranate) incorporated into the biocomposite film structure were collected from the Mediterranean and Central Anatolia regions of Türkiye.

2.2. Fruit Pretreatment

Pomegranate peels (Eskişehir) and hawthorn fruits (Antalya), selected as red fruit samples and obtained during their harvest season, were washed with distilled water, air-dried, and frozen overnight at −18 °C prior to lyophilization. The frozen samples were then ground into powder using a CryoMill grinder (Retsch) under a nitrogen atmosphere in 1-min intervals.

During the pretreatment process, hawthorn (H) and pomegranate peel (P) powders were mixed with distilled water at powder-to-water ratios of 1:4 (w/v) (Leaw, Kong and Pui, 2021) and 1:20 (w/v) (Ul-Islam et al., 2023), respectively, and stirred at approximately 50 °C under effective mixing conditions. The mixtures were subsequently placed in an ultrasonic bath for a total of 5 min in intermittent mode. The stirring durations were 24 h for hawthorn powder and 4 h for pomegranate peel powder. The resulting suspensions were centrifuged twice at 4500 rpm for 60 s. The obtained fruit powders were dried in a vacuum oven at 40 °C to remove excess moisture and stored in a desiccator until to use in the biocomposite film structure.

2.3. Production of Biocomposite Films

Biocomposite films were produced using four different formulations. An overview of the fabricated biocomposites and their preparation routes is provided in Table 1.

Table 1. Biocomposite Film Nomenclature and Formulation

Biocomposite Film	Formulation	Fruit Type
1BH	1	Hawthorn
2BP	2	Pomegranate Peel
3BH	3	Hawthorn
3BP	3	Pomegranate Peel
4BH	4	Hawthorn
4BP	4	Pomegranate Peel

Formulation 1

Chitosan and polyethylene glycol (PEG) were dissolved in a 2% acetic acid solution at approximately 50 °C (Pehlivanoğlu, 2021; Yılmaz, 2023). Fruit powder corresponding to 10% of the total polymer weight was dispersed in deionized water and stirred at room temperature for 2 h. The dispersion and glycerol were then added to the polymer solution and mixed using a magnetic stirrer at 45 °C until a homogeneous mixture was obtained. The mixture was subsequently stirred at room temperature for 18 h. Finally, it was treated in an ultrasonic bath for 10 min and cast into Petri dishes at equal volumes, followed by drying under ambient conditions.

Formulation 2

PVA and PEG were separately dissolved in distilled water. The polymer solutions were then combined at a weight ratio of 3:2 (PVA:PEG) (Çetinkaya, 2025). Fruit powder corresponding to 10% (w/w) of the total polymer content were weighed, first dispersed in a small amount of water, and then added to the polymer mixture. The solution was stirred until a homogeneous mixture was obtained. Glycerol corresponding to 10% (v/v) of the total solution volume was subsequently added. The resulting mixtures were cast into Petri dishes and allowed to dry at ambient temperature.

Formulation 3

Chitosan and PEG were dissolved in a 2% acetic acid solution at approximately 50 °C to obtain the first polymer mixture (Çetinkaya, 2025). PVA was separately dissolved in distilled water and added to the first polymer mixture at a weight ratio of 2:1 (PVA:polymer mixture), followed by stirring until a homogeneous solution was obtained. Fruit powder and glycerol corresponding to 10% (w/w) of the total polymer content were then added to the polymer mixture and stirred until homogeneous. The resulting mixtures were cast into Petri dishes and dried at ambient temperature.

Formulation 4

Polyvinyl alcohol (PVA) was dissolved in distilled water at approximately 90 °C under continuous stirring. Corn starch was dissolved in citric acid, and the starch solution was combined with the PVA solution, followed by stirring until a homogeneous mixture was obtained (Gerezgiher and Szabó, 2022). Fruit powder and glycerol, each corresponding to 10% of the total polymer weight, were incorporated into the polymer mixture and stirred until a homogeneous solution was achieved. The resulting mixtures were cast into Petri dishes and dried at 50 °C for 10 h, followed by curing at 120 °C for 1 h.

2.4. Liquid Exposure Stability Assessment

The potential direct exposure of biocomposite films to various liquids in packaging applications was considered, and their swelling and degradation behaviors were evaluated in acidic, basic, and alcoholic solutions. Aqueous acetic acid solutions (pH 4.5 and 6.5), a NaOH solution (pH 10), and a 70% (v/v) ethanol solution were used as the test media. From each biocomposite film, two specimens measuring 1×1 cm^2 were prepared, and their initial weights were recorded using an analytical balance. The samples were individually immersed in the respective solutions, and at designated time intervals, they were carefully removed, excess surface liquid was blotted with filter paper, and the samples were weighed. During the first day, measurements were taken every 30 min for the first 4 h. From the second day onwards, measurements were taken once daily, and the experiment was conducted over a total period of 7 days. The obtained weight changes were used to assess the swelling and degradation behaviors of the biocomposite films in different chemical environments.

2.5. Moisture Retention Capacity

The moisture retention capacity of the biocomposite films was determined using a gravimetric approach. Two specimens with dimensions of 1×1 cm^2 were cut from each biocomposite film, and their initial dry masses (m_0) were recorded. The samples were then placed in a desiccator containing deionized water and stored at room temperature to provide a humid environment. Relative humidity within the desiccator was monitored using a hygrometer. The masses of the specimens were measured at 24 h intervals. Moisture retention (%) was calculated according to Equation (1), where m_t represented the mass of the sample at time t and m0 corresponded to the initial dry mass.

$$Moisture\ retention\ capacity = \frac{(m_t - m_0)}{m_0}\ x\ 100 \qquad (1)$$

2.6. Determination of Swelling Degree

Specimens with dimensions of 1 × 1 cm^2 were prepared from the biocomposite films, and their initial dry masses (m_0) were determined using an analytical balance. The weighed samples were subsequently immersed in water-filled beakers. At predetermined time intervals, the biocomposite film specimens were periodically removed from the beakers, excess surface water was eliminated, and the samples were weighed. The swelling degree of the biocomposite films was calculated using Equation (2), where m_t represented the mass measured at the end of the immersion time t.

$$Swelling\ Degree = \frac{(m_t - m_0)}{m_0} \quad (2)$$

2.7. Biodegradability Analysis

The biocomposite films were cut into specimens with an area of 3 cm^2 and buried at a depth of 2 cm in soil-filled pots maintained under greenhouse conditions for 30 days. The masses of the biocomposite film specimens were recorded at 10-day intervals, and the corresponding mass losses were calculated using Equation (3). In this equation, W_0 represented the initial mass of the biocomposite film specimens, whereas W denoted the final mass measured at the end of the test period.

$$Mass\ Loss\ (\%) = \left[\frac{W_0 - W}{W_0}\right] x100 \quad (3)$$

2.8. Opacity Analysis

The UV transmittance performance of the biocomposite films was evaluated using an opacity test. The biocomposite films were cut into specimens with dimensions of 4 × 40 mm^2 and placed in the UV–Vis spectrophotometer cell. Absorbance spectra of the biocomposite films were recorded over the wavelength range of 250–850 nm. The opacity of the biocomposite films was calculated according to Equation (4), where A denoted the absorbance value at 600 nm (Zhao, Wang and Liu, 2022) and x represented the thickness of the biocomposite film.

$$Opacity = \frac{A}{x} \quad (4)$$

2.9. Morphological Assessment by Optical Microscopy

Optical microscopy images were used to qualitatively and quantitatively evaluate the distribution at the reinforcement–matrix interface. The prepared biocomposite film specimens were placed on microscope slides and covered with a coverslip to ensure a flat and stable surface. Imaging was performed using a Leica S8apo stereomicroscope in bright-field mode at 100× magnification, providing sufficient contrast and resolution to assess reinforcement dispersion, interfacial heterogeneity, the morphology of the matrix phase surrounding the reinforcement materials, and potential cross-linking regions. When necessary, contrast and illumination settings were optimized for clear visualization.

2.10. Contact Angle Measurement

Contact angle measurements of the biocomposite film surfaces were performed using an Attension Theta optical tensiometer. A droplet of deionized water was deposited onto the biocomposite film surface using the instrument's syringe system. The interaction between the water droplet and the surface was recorded by a high-resolution camera, and the contact angles were subsequently determined.

2.11. Scanning Electron Microscopy (SEM) Analysis

The morphological characterization of the biocomposite films was conducted at the Eskişehir Osmangazi University (ESOGU) Research and Application Center (ARUM) using a JEOL JSM-5600LV SEM. Micrographs were acquired at an accelerating voltage of 10.0 kV and a magnification of 1000×.

2.12. Tensile Strength Behavior of Biocomposite Films

The mechanical properties of the biocomposite films were evaluated using a ZwickRoell tensile testing machine. From each biocomposite film, specimens measuring 13×15 mm^2 were carefully cut with uniform thickness. Tensile tests were performed with a load limit of 500 N and a crosshead speed of 1 mm min^{-1}. During the tests, the elongation at break (μm) and the corresponding force at failure (N) were recorded for each specimen.

2.13. Antibacterial Activity Evaluation via Disk Diffusion Method

The biocomposite films were initially examined for potential contamination, and only the samples confirmed to meet aseptic conditions were subsequently subjected to antibacterial evaluation using the disk diffusion method. The antimicrobial activities of the samples were assessed against Staphylococcus aureus and Escherichia coli using the disk diffusion assay. The density of the bacterial cultures was spectrophotometrically measured at a wavelength of 600 nm (OD_{600}) to standardize the cell concentration. Bacterial suspensions adjusted to an optical density of 1.0 were considered to correspond to approximately 8 × 10^8 CFU/mL, based on correlations reported in the literature (Christensen et al., 1985). Aliquots of 100 μL (≈10^8 CFU) from the prepared bacterial suspensions were evenly spread onto the

surface of solid LB agar media. Test samples (3BH and 3BP), along with negative and positive control disks, were placed at equal distances on the inoculated Petri dishes. Sterile disks impregnated with 20 µL of ciprofloxacin solution (1 mg/mL) were used as the positive control, while sterile disks loaded with 20 µL of LB broth served as the negative control. All Petri dishes were incubated at 37°C for 16–18 h under conditions suitable for bacterial growth. Following incubation, the inhibition zones formed around the samples were digitally imaged, and the inhibition diameters were quantified using ImageJ software. The measured inhibition zones were used to evaluate the antibacterial effectiveness of the biocomposite film samples.

3. Results & Discussions

3.1. Liquid Exposure Stability

The durability of the biocomposite films against different solutions was assessed through mass loss measurements, and the total mass loss percentages in each solvent medium were summarized after seven days of exposure in Table 2.

Table 2. Mass Loss of Biocomposite Films in Various Solvent Media

	1 (pH =4.5)	2 (pH =6.5)	3 (pH =10)	4 (Alcohol) (70%)
1BH	1.1	4.8	1.8	4.2
2BP	-	-	10.4	17.1
3BH	12.6	11.3	18.4	10.5
3BP	12.0	6.5	22.5	11.9
4BH	16.5	6.5	6.6	14.7
4BP	3.9	4.6	5.0	2.8

The behavior of the biocomposite films in different pH environments and alcoholic solutions indicated that pH is a key factor affecting material stability. Observable degradation began in the 2NB sample at 18 h in pH 4.5 and at 24 h in pH 6.5. Samples 1BH and 4BP remained more stable than the others in all tested media. These findings highlight the importance of matching biocomposite films performance to the pH of the intended application environment.

Overall, the biocomposite films remained more stable and underwent less degradation in the second solvent medium, which was close to neutral pH. All polymers incorporated into the composite structure were structurally more prone to degradation under acidic conditions. In acidic environments, the amino groups of chitosan became protonated, leading to an increase in the intermolecular distance between polymer chains. This expansion facilitated greater solvent penetration between the chains, thereby accelerating degradation processes (Dash, Chiellini, Ottenbrite and Chiellini, 2011; Rinaudo, 2006)

Poly(vinyl alcohol) (PVA) contains a high density of hydroxyl (–OH) groups, which were susceptible to degradation under acidic conditions. Similarly, the glycosidic bonds present in starch were readily hydrolyzed in acidic media (Bertoft, 2017). In addition, the ether linkages in the polyethylene glycol (PEG) structure were known to undergo hydrolysis in strongly acidic environments (Ewald et al., 2020). The pronounced mass loss observed for the 4BH biocomposite film under acidic conditions confirmed that the materials experienced actual chemical degradation in this medium, and that the observed loss was directly associated with degradation phenomena.

However, an examination of the experimental results (Table 2) revealed that the remaining biocomposite films exhibited higher mass loss in alkaline media compared to acidic conditions. This behavior could be attributed to several factors. Deprotonation of the amino groups in the chitosan structure resulted in the disruption of ionic interactions within the film matrix. Consequently, the polymer network loosened, allowing plant additives embedded in the structure to be released into the solvent medium, leading to mass loss. This type of loss was primarily physical in nature and was not directly related to polymer degradation.

A similar effect may have occurred in the other polymers due to the weakening of hydroxyl-mediated interactions within the structure. Furthermore, pomegranate peel and hawthorn powder inherently possess acidic characteristics owing to their phenolic acid content and therefore exhibited a higher tendency to dissolve in alkaline environments (El-Hamamsy and El-Khamissi, 2020; Kara, Kara and Dinç, 2025). This behavior likely accelerated the migration of plant ingredients into the solvent under alkaline conditions, resulting in increased mass loss. In summary, the mass loss observed in alkaline media could be associated not only with degradation but also with dissolution-related processes.

Under alcoholic conditions, 2BP displayed a markedly higher tendency toward dissolution than the other biocomposite films. This behavior was primarily attributed to the presence of polyethylene glycol (PEG) in the 2BP structure, which is highly soluble in alcohol. Moreover, the absence of polymers with limited alcohol solubility, such as chitosan and starch (Rinaudo, 2006; Tester, Karkalas and Qi, 2004), may have rendered this formulation more susceptible to the solvent medium. The comparatively higher resistance of the remaining biocomposite films to the alcoholic environment can be explained by the poor solubility of their constituent

polymers in alcohol, leading predominantly to swelling rather than dissolution.

3.2. Moisture Retention Capacity

The moisture retention percentages of the biocomposite films are presented in Table 3. All biocomposite films exhibited moisture retention below 10%. Among all samples, 2BP showed slightly higher moisture retention compared to the others. Additionally, 1BH displayed visible signs of degradation by the second day, which may be related to the lower crosslinking density in this sample. 4BH and 4BP consistently demonstrated the lowest moisture retention percentages, a result associated with more effective crosslinking among the polymers constituting the composite structure. Crosslinking has been reported to restrict the availability of hydrophilic functional groups in chitosan, thereby hindering water vapor adsorption into the film matrix (Berger et al., 2004). Despite their lower moisture retention, 4BH and 4BP may be more suitable for applications in humid environments due to their slower degradation behavior. Furthermore, the moisture retention capacities of the biocomposite films were found to be comparable to or lower than those reported for biocomposite films with similar compositions containing natural fillers (Pavlovic, Valzania, and Minak, 2025).

Table 3. Moisture Retention Percentages of The Biocomposite Films

	t=24 h	t=48 h
1BH	4,07	-
2BP	6,21	7,97
3BH	5,66	5,74
3BP	4,42	4,14
4BH	0,29	0,41
4BP	0,40	0,72

3.3. Swelling Degree

The swelling test results were presented in Figure 1. The findings indicated that the swelling degrees of all biocomposite films remained below 10. Among the samples, 2BP exhibited the highest water uptake capacity, whereas 4BH and 4BP showed comparatively lower swelling degrees, demonstrating a high resistance to dimensional expansion. The ranking of swelling degrees was consistent with the moisture retention results. No reliable swelling data could be obtained for 1BH due to its excessively rapid swelling behavior upon immersion in water. This limitation should be considered when interpreting the comparative swelling behavior.

Swelling behavior was primarily governed by the hydrophilic/hydrophobic nature of the material as well as the degree of crosslinking within the polymer network. Crosslinking, defined as the formation of additional interchain bonds (Ahmed, 2015), reduced the free volume between polymer chains and limited solvent diffusion into the matrix. Consequently, the lower swelling observed in 4BH and 4BP suggested a more densely crosslinked structure, leading to restricted chain mobility and enhanced structural stability. Furthermore, crosslinking-induced masking of hydrophilic functional groups in chitosan may have further reduced water affinity and suppressed swelling.

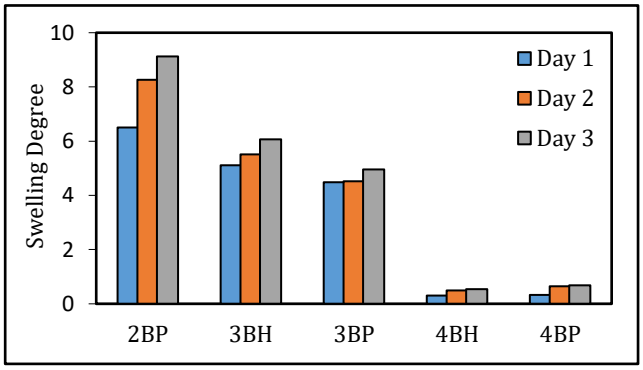


Figure 1. Swelling Degrees of Biocomposite Films in Water

3.4. Biodegradation Performance

Time-dependent mass changes of soil-buried biocomposite films were presented in Figure 2. Complete degradation of 1BH occurred within 10 days. In contrast, the fourth formulation biocomposite films (4BH and 4BP) showed the slowest degradation kinetics, with less than 10% mass loss recorded after 20 days. Overall, the degradation rates of the biocomposite films were substantially faster than those reported for conventional plastic alternatives used in packaging applications (Afshar, Boldrin, Astrup, Daugaard and Hartmann, 2024; Erkul and Uçaroğlu, 2023; Niu et al., 2024).

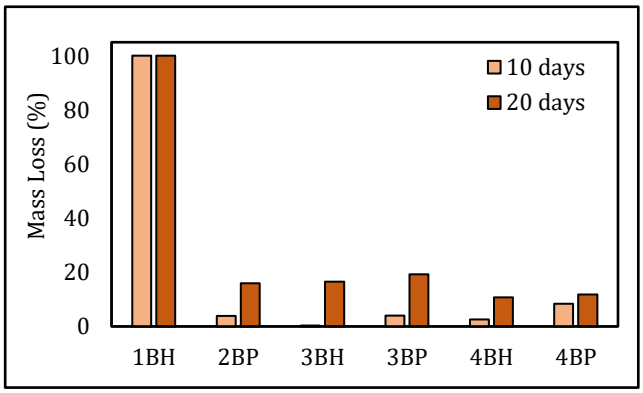


Figure 2. Mass Loss of Biocomposite Films During Soil Burial

3.5. Opacity Evaluation

The opacity values of the biocomposite films were calculated using Equation (4) for each sample and were presented in Figure 3. Opacity performance was inversely correlated with material transmittance, where lower transmittance and correspondingly higher opacity were required for effective UV shielding (Fan and Picchioni, 2020).

Films with lower transparency are often associated with more effective UV protection, as reported in previous studies (Kameneva et al., 2020). Based on the evaluation of light transmittance through opacity values, all investigated biocomposite films exhibited opacity values higher than that of 2BP (0.88), while the other samples showed opacity values ranging between 1.2 and 1.7. These findings indicated that the prepared biocomposite films possessed low light transmittance together with relatively high opacity, suggesting that they could act as UV barriers to a certain extent and could be considered suitable candidates for packaging applications.

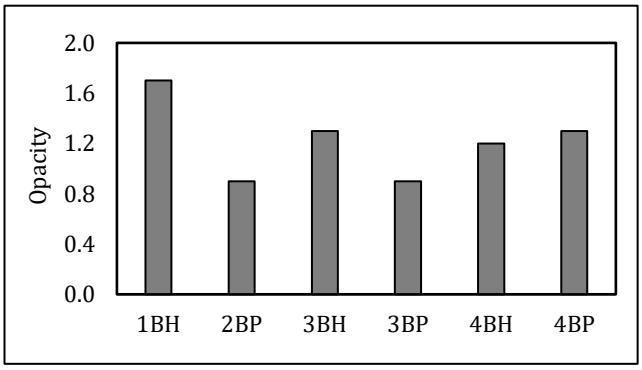


Figure 3. Opacity Values Related to UV Shielding of Biocomposite Films

3.6. Optical Microscopy Observations

The optical microscopy images of biocomposite films are shown in Figure 4. The micrographs provided evidence of crosslinking and fruit additive–matrix interfacial interactions. The biocomposite films prepared using the third formulation (3BH and 3BP) exhibited a more homogeneous distribution of the reinforcing components within the polymer network and stronger interfacial interactions compared to the other formulations.

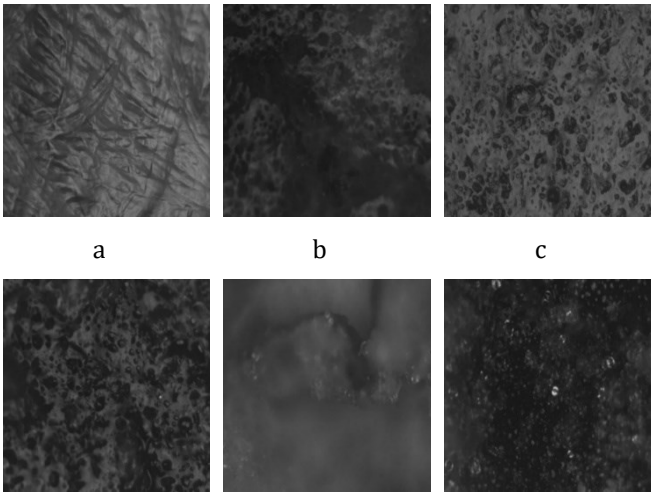
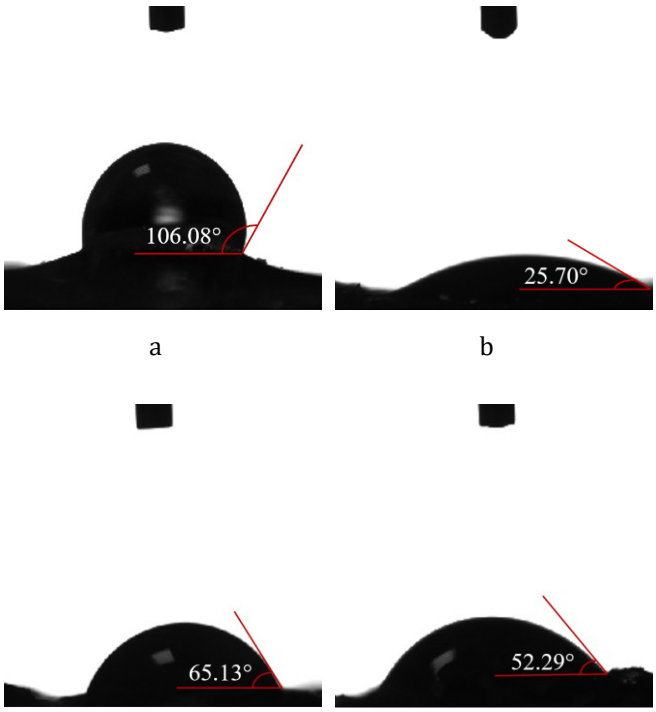


Figure 4. Optical Microscopy Images of The Biocomposite Films: A) 1BH, B) 2BP, C) 3BH, D) 3BP, E) 4BH, F) 4BP

3.7. Contact Angle Measurements and Surface Wettability

In solids with high surface energy, liquids form small contact angles (<90°) with the surface, reflecting hydrophilic behavior, whereas low-surface-energy solids exhibit large contact angles (>90°), characteristic of hydrophobic surfaces (Hamraoui, 2025; Yılmaz et al., 2025). A decrease in contact angle is associated with an increase in surface energy, adhesion, and wettability, whereas an increase in contact angle corresponds to a reduction in surface energy, wettability, and adhesion (Adamson, A.W., 1990).

The interactions between water droplets and the biocomposite film surfaces, together with the corresponding contact angle values, are presented in Figure 5. The results indicated that the 2BP biocomposite films exhibited more hydrophilic characteristics compared to the other samples, and this finding was consistent with the moisture retention and swelling degree results. The contact angle of the 1BH biocomposite film was found to be approximately 106°. This relatively high value was attributed to the irregular surface morphology observed in optical microscopy images, characterized by pits and protrusions. It was considered that this non-uniform surface structure significantly affected the contact angle measurements.



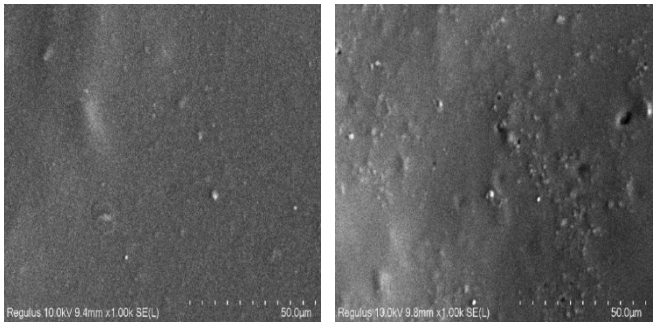
a b

c d

Figure 5. Surface Wettability and Contact Angles of The Biocomposite Films: a) 1BH, b) 2BP, c) 3BH, d) 3BP

3.8. SEM-Based Surface Morphology

SEM images were acquired to further examine the 3BH and 3BP biocomposite films, which exhibited homogeneous dispersion in optical microscopy, and are presented in Figure 6. The raised features observed across the material surface in the SEM micrographs originated from the reinforcing components, confirming their uniform distribution within the polymer matrix. In addition, the SEM observations revealed smooth surface morphologies and indicated that the biocomposite films were microstructurally stable.

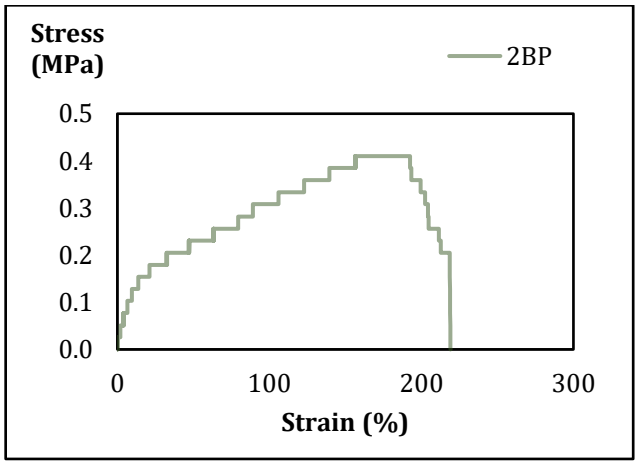


a b

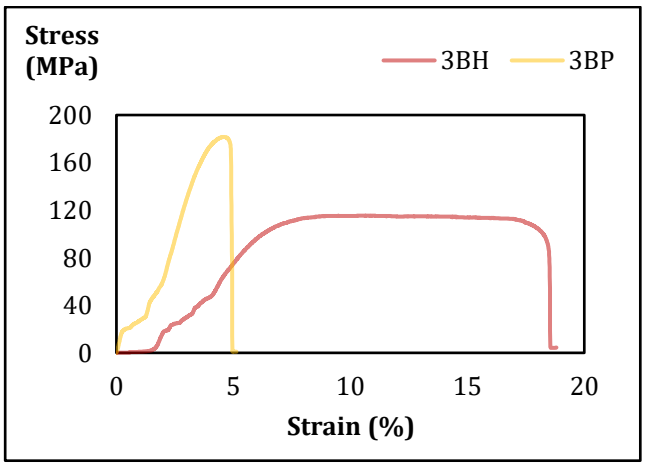
Figure 6. SEM Micrographs of The Biocomposite Films: A) 3BH, B) 3BP

3.9. Tensile Stress–Strain Behavior

The tensile stress–strain curves of the biocomposite films prepared using the second and third formulations are presented in Figure 7. The tests revealed that 3BH and 3BP exhibited higher tensile strength compared with 2BP. The biocomposite films from the second formulation contained PVA and PEG polymers, which can form only hydrogen bonds without crosslinking, and the incorporation of PEG into the PVA matrix provided additional flexibility (Falqi, Bin-Dahman, Hussain, and Al-Harthi, 2018). In contrast, the third formulation included chitosan, PEG, and PVA, allowing crosslinking between the polymer chains. This crosslinking enhanced the mechanical strength of the biocomposite films.



a



b

Figure 7. Tensile stress–strain curves of the biocomposite films: a) 2BP, b) 3BH and 3BP

Biocomposite films containing natural fibers and similar biopolymers have reported tensile strengths ranging from 1.0 to 1.5 MPa (Mohammed and El-Sayed, 2023). However, the tensile strength of the 2BP biocomposite film was found to be 0.4 MPa, which

remained below the reported literature range. This reduction in mechanical performance was attributed to the non-uniform dispersion of reinforcing fruit additives within the polymer matrix and their partial agglomeration in certain regions. The third-formulation biocomposite films displayed reported tensile strengths between 73 and 147 MPa (Rihayat and Hasnah, 2020). Within this context, 3BH fell within this range, while 3BP demonstrated higher mechanical strength, likely due to the uniform distribution of reinforcing components, strong inter-component compatibility, and effective crosslinking.

Additionally, 3BH, containing hawthorn, exhibited greater elongation and flexibility compared with those containing pomegranate peel. It was reported that pomegranate peel were lignocellulosic with a relatively high lignin content; lignin was associated with increased fiber stiffness and brittleness, while fibers derived from softer plant tissues such as fruit peels were generally finer in structure (Eleutério, Trota, Meirelles and Vasconcelos, 2025; Hasnaoui, Wathelet and Jiménez-Araujo, 2014). This difference likely contributed to the higher strain values and enhanced elongation at break observed in the hawthorn-containing biocomposite film. Swelling tests further indicated that the third-formulation biocomposite films exhibited maximum swelling of 4–6%, while the second-formulation biocomposite films reached approximately 9%, which reflected stronger crosslinking in the third-formulation samples. Consequently, the lower degree of crosslinking in 2BP explained their reduced tensile strength.

3.10. Antibacterial Performance of Biocomposite Films

The disk diffusion assay demonstrated that the 3BP biocomposite film exhibited antibacterial activity against S. aureus and E. coli by forming distinct inhibition zones (Figure 8). The inhibition area, defined as the difference between the total inhibited region and the biocomposite film area, measured 3 mm^2 against S. aureus, which was 2.14 times larger than that observed against E. coli (1.4 mm^2). These findings suggested that 3BP was more effective against Gram-positive bacteria and that differences in bacterial cell wall architecture may have influenced the antibacterial activity of the biocomposite film. This observation was consistent with previous studies reporting that Gram-negative bacteria exhibit greater resistance to various polymers compared with Gram-positive bacteria (Cox and Wright, 2013; Gupta et al., 2020; Silhavy, Kahne and Walker, 2010). While S. aureus is a Gram-positive bacterium characterized by a thick peptidoglycan layer, E. coli is a Gram-negative bacterium possessing a more complex cell wall structure with an outer membrane composed of lipopolysaccharides (Ruhal and Kataria, 2021). This outer membrane limits the penetration of macromolecules, including antimicrobial agents, thereby contributing to bacterial resistance mechanisms (Smola-Dmochowska et al., 2023), which explains the smaller inhibition zone observed for E. coli.

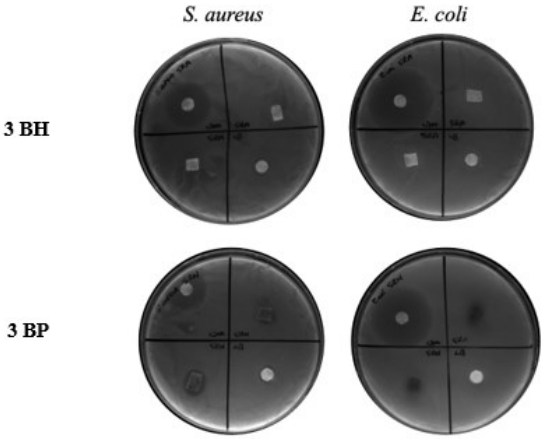


Figure 8. Disk Diffusion Assay of 3BH and 3BP Biocomposite Films Against *S. aureus* and *E. coli*

In contrast to 3BP, 3BH did not produce inhibition zones against either bacterial strain. Both biocomposite films were prepared using the same formulation, with the only difference being the type of plant incorporated into the structure. Accordingly, the type of plant used could be considered an important factor influencing the antibacterial properties of biocomposite films. Pomegranate peel was reported to contain hydrolysable tannins such as gallic acid, ellagic acid, and punicalagin (Singh, Singh, Kaur, and Singh, 2018). These compounds may exert antibacterial effects by interacting with bacterial cell membranes, protein structures, or enzymatic activities. In contrast, hawthorn was reported to be richer in flavonoids and phenolic acids, which are more commonly associated with antioxidant and anti-inflammatory activities (Alirezalu et al., 2020). Therefore, the relatively stronger antibacterial potential of the compounds presents in pomegranate peel compared to those in hawthorn could partly explain the formation of inhibition zones in 3BP, whereas no inhibition was observed for 3BH.

4. Conclusion

This work presented the preparation and comprehensive evaluation of bio-based biocomposite films using chitosan, polyethylene glycol (PEG), and polyvinyl alcohol (PVA) biopolymers, together with locally sourced hawthorn (*Crataegus* spp.) and pomegranate (*Punica granatum* L.), as an environmentally friendly alternative to petroleum-based packaging materials. The results indicated biocomposite films exhibiting properties appropriate

for potential packaging applications. Chemical stability tests showed that the films maintained structural integrity and high stability after exposure to acidic, basic, and alcoholic solutions for seven days. Moisture retention and swelling analyses demonstrated strong crosslinking among polymer chains, particularly in the second, third, and fourth biocomposite film formulations. Soil burial tests validated degradation within a much shorter period compared with commercial plastic packaging materials, indicating high biodegradability potential. Opacity measurements revealed superior ultraviolet (UV) protection for the 3BP biocomposite film compared with the other formulations. Contact angle measurements illustrated incorporation of chitosan modifying surface characteristics and shifting the materials toward more hydrophobic behavior. Optical microscopy and SEM analyses confirmed homogeneous dispersion of the reinforcing materials within the polymer matrix. In addition, the type of plant-derived additive significantly affected the functional properties of the developed biocomposite films. The biocomposite film containing hawthorn (3BH) exhibited greater flexibility and elongation, whereas the pomegranate peel-containing biocomposite film (3BP) showed higher stress resistance and detectable antibacterial activity, particularly against S. aureus. These differences may be associated with the structural characteristics of the additives, since pomegranate peel is rich in lignocellulosic structures and bioactive phenolic compounds, while hawthorn may contribute to a softer and more flexible film structure. Overall, these supported revealed natural ingredient-reinforced biocomposite films representing a promising approach for the development of environmentally friendly, functional, and biodegradable packaging materials.

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Author Contributions

In this study, Fırat YILMAZ was responsible for the literature review, manuscript writing, and visualization; Demet TOPALOĞLU YAZICI contributed to the literature review, methodology development, and supervision; Asu Hümeyra DOĞU, Aybeniz SAVAŞ, İrem YEŞİL and Semiha TAŞKIN were involved in the literature review and carried out the experiments.

Conflict of Interest

The authors declare no conflict of interest.

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