Putra *et al.* (2026) Vol. 14 No. 1: 83-90

Chimica et Natura Acta

p-ISSN: 2355-0864 e-ISSN: 2541-2574

Homepage: http://jurnal.unpad.ac.id/jcena

Influence of Polyvinyl Alcohol Content on Starch-Based Bioplastic Reinforced with Chitosan and Microcrystalline Cellulose

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DOI: https://doi.org/10.24198/cna.v14.n1.70771

Article history: Received 12 March 2026 | Revised 24 April 2026 | Accepted 10 May 2026

Abstract: Starch-based bioplastics are promising sustainable alternatives to petroleum-based plastics; however, their limited mechanical strength and stability restrict broader packaging applications. This study investigates the influence of polyvinyl alcohol (PVA) content on the structural and functional properties of starch-based bioplastic films reinforced with chitosan and microcrystalline cellulose (MCC). Films prepared by solution casting with varying PVA compositions were characterized in terms of density, mechanical properties, thermal stability, biodegradation behavior, antibacterial activity, and structural features. Increasing PVA content produced denser film structures with improved tensile strength and thermal stability, while elongation at break reached its maximum at the intermediate formulation, indicating a strength–ductility trade-off. The control film exhibited antibacterial inhibition zones of 12.6 mm against Staphylococcus aureus and 8.8 mm against Escherichia coli, with activity decreasing at higher PVA content. These findings demonstrate that adjusting PVA composition effectively tailors the multifunctional performance of starch-based bioplastic films for sustainable packaging applications.

Keywords: bioplastics, potato peel starch, polyvinyl alcohol, microcrystalline cellulose, antibacterial activity

Abstrak: *Bioplastik berbasis pati merupakan alternatif berkelanjutan yang menjanjikan untuk menggantikan plastik berbasis petroleum, namun keterbatasan kekuatan mekanik dan stabilitasnya masih membatasi penerapannya pada kemasan. Penelitian ini mengkaji pengaruh variasi kandungan polyvinyl alcohol (PVA) terhadap sifat struktural dan fungsional film bioplastik berbasis pati yang diperkuat dengan kitosan dan mikrokristalin selulosa (MCC). Film disintesis melalui metode solution casting dengan variasi komposisi PVA dan dikarakterisasi melalui pengukuran densitas, sifat mekanik, stabilitas termal, biodegradasi, aktivitas antibakteri, serta analisis struktur menggunakan FTIR, XRD, dan SEM. Peningkatan kandungan PVA menghasilkan struktur film yang lebih padat serta meningkatkan kekuatan tarik dan stabilitas termal, sedangkan nilai elongation at break mencapai maksimum pada komposisi menengah yang menunjukkan adanya trade-off antara kekuatan dan keuletan material. Film kontrol menunjukkan zona hambat antibakteri sebesar 12,6 mm terhadap Staphylococcus aureus dan 8,8 mm terhadap Escherichia coli, dengan aktivitas yang menurun pada kandungan PVA yang lebih tinggi. Hasil ini menunjukkan bahwa pengaturan komposisi PVA merupakan strategi efektif untuk mengendalikan sifat mekanik, struktural, dan fungsional film bioplastik berbasis pati untuk aplikasi kemasan berkelanjutan.*

Kata kunci: *bioplastik, pati kulit kentang, polyvinyl alcohol, mikrokristalin selulosa, aktivitas antibakteri*

INTRODUCTION

The extensive use of petroleum-based plastics has raised significant environmental and sustainability concerns due to their persistence in natural ecosystems and contribution to pollution and microplastic formation (Banu & Sharmila 2023). Plastic leakage into soil and aquatic environments, particularly marine ecosystems, has intensified the search for biodegradable and bio-based alternatives to reduce long-term environmental impacts (Bartolucci *et al.* 2023). Within circular economy frameworks, bioplastics are increasingly explored as materials that may reduce reliance on fossil resources and potentially lower greenhouse gas emissions when combined with appropriate end-of-life management strategies (Keith *et al.* 2024). These environmental challenges highlight the urgent need to develop sustainable materials that reduce long-term plastic accumulation.

Among renewable polymers, starch has attracted considerable attention due to its abundance, renewability, and relatively low cost (Shrestha *et al.* 2023). Starch derived from agro-industrial residues such as potato peel is particularly promising because it enables the valorization of food-processing waste while supporting circular economy principles (Shah *et al.* 2021). Starch-based materials can exhibit favorable biodegradation behavior under suitable environmental conditions, making them attractive candidates for sustainable packaging applications (Serrano-Aguirre & Prieto 2024).

However, native starch films still suffer from inherent limitations such as low mechanical strength, brittle behavior, and high moisture sensitivity, which restrict their direct application in packaging systems (Rosenboom *et al.* 2022). To overcome these limitations, various modification strategies have been proposed, including polymer blending, plasticization, crosslinking, and filler reinforcement. Polyvinyl alcohol (PVA) is commonly blended with starch because its hydroxyl groups can form hydrogen bonds with starch molecules, improving interfacial adhesion and mechanical performance in composite films (Diyana *et al.* 2021). In potato peel starch systems, the addition of PVA has been shown to enhance structural integrity and tensile properties when appropriate formulation and processing conditions are applied (Charles *et al.* 2022). Chitosan and microcrystalline cellulose (MCC) are also frequently incorporated as functional additives, where chitosan provides antimicrobial functionality and MCC contributes mechanical reinforcement within the polymer matrix (Falua *et al.* 2025; Tan *et al.* 2022). Combining polymer blending and filler reinforcement, therefore, represents a promising strategy for improving the performance of starch-based bioplastics.

Despite these advances, systematic studies investigating how variations in PVA composition influence the multifunctional performance of potato-peel-starch bioplastics reinforced with chitosan and MCC remain limited. Most previous studies focus on individual performance aspects, whereas comprehensive evaluations linking PVA variation to structural, mechanical, thermal, biodegradation, and antibacterial characteristics remain scarce (Diyana *et al.* 2021; Tan *et al.* 2022). Understanding these relationships is essential for designing biodegradable materials with balanced structural and functional properties for sustainable packaging applications. Therefore, this study aims to investigate the influence of PVA content on starch-based bioplastic films derived from potato peel starch and reinforced with chitosan and microcrystalline cellulose. The films were prepared by solution casting and characterized for density, mechanical performance, thermal stability, biodegradation behavior, antibacterial activity, and structural properties.

MATERIALS AND METHOD

Materials

Potato peels (*Solanum tuberosum* L.) were obtained from a traditional market in Malang, Indonesia. Polyvinyl alcohol (PVA; CAS 9002-89-5; degree of hydrolysis 86.5–89 mol%; Mn ≈ 160 kDa) and chitosan (CAS 9012-76-4; degree of deacetylation ≥ 75%; Mn 3.8–20 kDa) were purchased from HiMedia Laboratories (India). Microcrystalline cellulose (MCC; CAS 9004-34-6; particle size ≈ 20 μm), sodium hydroxide (NaOH, ≥ 99%; CAS 1310-73-2), hydrogen peroxide (H_2O_2, 30%; CAS 7722-84-1), glacial acetic acid (CAS 64-19-7), glycerol (≥ 99.5%; CAS 56-81-5), and gelatin (analytical grade) were purchased from Merck (Germany). Distilled water was used in all experiments.

Potato Peel Starch Preparation

Potato peels were washed and sun-dried under direct sunlight until completely dehydrated. The dried peels were ground with a grinder and then sieved through a 120-mesh sieve to obtain fine powder. Starch extraction was performed via alkaline treatment using 15% (w/v) NaOH solution with a solid-to-liquid ratio of 1:12 (w/v) at 100°C under constant stirring (240 rpm) for 1 h (Afifah *et al.* 2022). The resulting slurry was washed with distilled water until a neutral pH (~7) was reached. Subsequently, a bleaching step was carried out using 10% (w/v) H_2O_2 for 2 h. The product was oven-dried for 8 h, and sieved (120 mesh) to obtain fine starch powder.

Bioplastic Film Synthesis

PVA (0, 5, 7.5, and 10 g) was dissolved in 100 mL of distilled water at 80°C under continuous stirring to obtain four formulations, designated P0, P5, P7.5, and P10. In all formulations, potato peel starch, MCC, and chitosan were maintained at a constant amount of 1 g each and subsequently added to the PVA solution. Separately, 2.5 g of gelatin was dissolved in 15 mL of distilled water at 50°C. Glacial acetic acid (10 mL) was then added to the gelatin solution, after which the mixture was incorporated into the bioplastic solution. Glycerol (0.3 mL) was added as a plasticizer. The mixture was magnetically stirred for 30 min to achieve homogenization, and then cast into Petri dishes and dried in an oven at 45°C for 16 h. The resulting films were peeled off and stored for further characterization.

Characterizations

Film density was determined from the mass-to-volume ratio using an analytical balance (KERN) and a digital caliper (Mitutoyo). Biodegradation was evaluated using the soil burial method, with measurements conducted at 0, 7, 14, and 21 days. Tensile testing was conducted using a Universal Testing Machine (Instron 5567, USA) following ASTM D882, with a gauge length of 50 mm and a crosshead speed of 10 mm min⁻¹. Thermal stability was analyzed using a simultaneous thermal analyzer (Linseis STA PT1600) from 40–400°C in air following ASTM E1131. Functional groups were identified by FTIR spectroscopy (Shimadzu IRSpirit-T, ATR mode, 4000–400 cm⁻¹). The crystalline structure of the films was analyzed by X-ray diffraction (PANalytical X'Pert PRO, Cu Kα radiation, 2θ = 10–65°), while surface morphology was observed using SEM (Hitachi TM4000PlusII). Antibacterial activity against *Staphylococcus aureus* and *Escherichia coli* was assessed using a disc diffusion assay on Mueller–Hinton agar, followed by incubation at 37 °C for 24 h.

RESULTS AND DISCUSSION

Density and Biodegradation

As shown in Figure 1(a), the density of the bioplastic films increased progressively with higher PVA content. The control sample (P0) displayed the lowest density, while the P10 formulation exhibited the highest value, suggesting the development of a more compact film network. This behavior is mainly associated with stronger hydrogen-bond interactions and tighter molecular packing between starch and PVA chains, resulting in a more compact molecular arrangement within the polymer network (Nugroho *et al*. 2024). Studies on starch–PVA composite films indicate that intermolecular interactions between polymer chains enhance the structural cohesion of the film matrix (Janik *et al*. 2018).

The biodegradation behavior obtained from the soil burial test is presented in Figure 1(b). The starch-rich control film exhibited the highest weight loss, indicating faster degradation compared with films containing higher PVA fractions. Increasing PVA content generally reduced the degradation rate, suggesting improved structural stability of the films in soil environments. Starch-based polymers degrade rapidly due to their hydrophilic nature and susceptibility to microbial attack (Enwere *et al*. 2024). The incorporation of PVA can slow this process by strengthening the film network and reducing excessive water uptake (Ranote *et al*. 2024).

FTIR Analysis

Figure 2(a) presents the infrared spectra recorded for the prepared bioplastic films. The spectrum shows an extended band around 3600–3200 cm⁻¹, corresponding to O–H stretching modes originating from hydroxyl functionalities of starch, PVA, and chitosan. As the PVA content increases, this band becomes slightly broader and shifts marginally, suggesting stronger intermolecular hydrogen-bond interactions within the film network. Previous studies

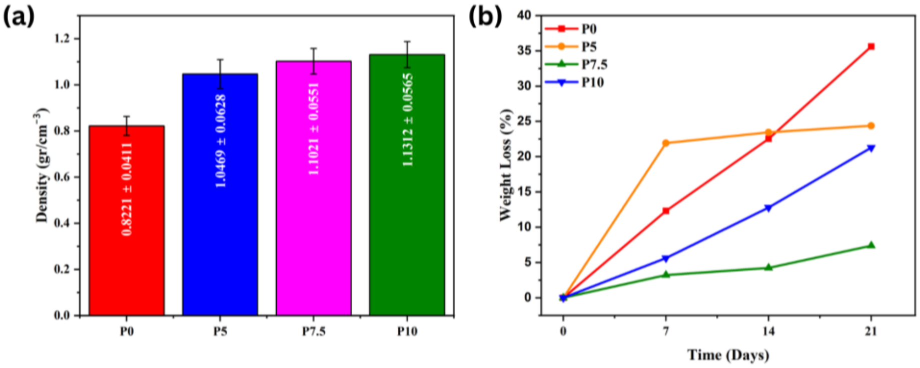

Figure 1. (a) Density and (b) biodegradation of the bioplastics

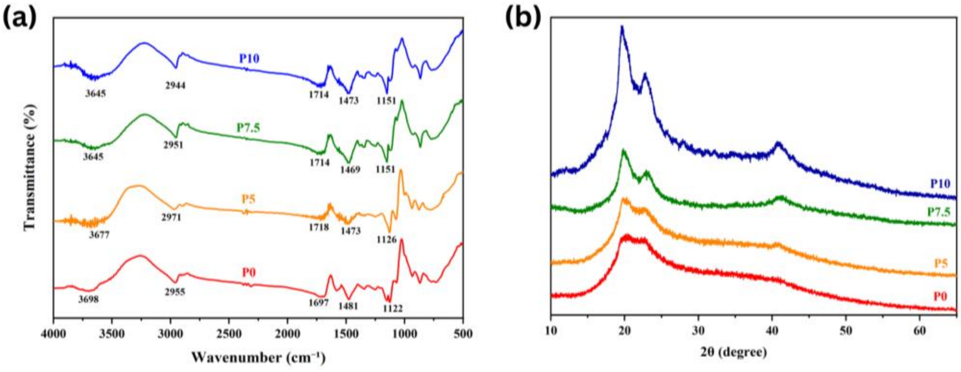

Figure 1. (a) FTIR and (b) XRD of the bioplastics.

on starch–PVA composites have reported comparable spectral behavior, indicating that hydrogen bonding plays an important role in improving compatibility between polymer chains (Ahmed *et al*. 2025).

Characteristic bands observed at 1151–1122 cm⁻¹ are attributed to C–O stretching vibrations, while peaks around 1080–1020 cm⁻¹ correspond to C–O–C glycosidic linkages of polysaccharide backbone. In the fingerprint region (1000–600 cm⁻¹), multiple overlapping peaks reflect complex vibrational modes associated with starch and chitosan structures, further confirming the presence of these components in the composite films (Castro *et al*. 2023). In addition, a band around 2971–2944 cm⁻¹ assigned to C-H stretching vibrations shows a slight shift toward lower wavenumbers with increasing PVA content, indicating enhanced intermolecular interactions between polymer chains. A weak absorption observed at approximately 1718–1697 cm⁻¹ is associated with residual acetate groups derived from PVA, confirming its incorporation into the composite structure. The absence of additional characteristic peaks suggests that the interactions among the components mainly occur through physical interactions rather than the formation of new covalent bonds (Wang *et al*. 2024).

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XRD Analysis

The X-ray diffraction profiles of the prepared bioplastic films are shown in Figure 2(b). All formulations show a distinct diffraction feature at approximately 2θ = 19–20°, which is typically associated with the semi-crystalline organization found in starch–PVA systems. The degree of crystallinity appears to vary with PVA composition, showing a slight increase at moderate PVA content, followed by a decrease at the intermediate formulation (P7.5), and reaching the highest level at the highest PVA fraction. Such variations are typical in polymer blends where intermediate compositions temporarily disrupt ordered domains before new crystalline arrangements develop at higher polymer fractions (Ningrum *et al*. 2025). The higher crystallinity observed at elevated PVA content indicates improved chain organization and stronger molecular cohesion within the films. Changes in crystallinity are known to influence mechanical behavior because higher crystallinity generally increases stiffness, whereas lower crystallinity permits greater chain mobility (Abe *et al*. 2024).

Mechanical Properties

The mechanical performance of the bioplastic films is presented in Figure 3, showing the variation in tensile strength and elongation at break. Increasing PVA content resulted in higher tensile strength, with the P10 formulation exhibiting the highest value, suggesting improved load-bearing capacity of the films. This behavior is mainly attributed to stronger intermolecular interactions between starch and PVA chains, which enhance cohesion within the polymer network. In addition, the semi-crystalline nature and linear chain structure of PVA contribute to improved stress transfer and reduced chain slippage, further enhancing the tensile strength of the composite films. Similar strengthening effects have been reported in starch–PVA systems where hydrogen bonding improves the mechanical performance of biopolymer matrices (Nugroho *et al*. 2024).

In contrast, elongation at break exhibited a different trend, with the intermediate formulation (P7.5) showing the highest ductility among the samples. This behavior may be related to its relatively lower crystallinity, which increases chain mobility and facilitates plastic deformation during tensile loading. Conversely, stronger hydrogen bonding and higher crystallinity at elevated PVA content restrict chain movement and reduce flexibility. This behavior reflects the well-known strength–ductility trade-off in polymer composites, where tighter chain packing enhances tensile strength while limiting elongation capacity (Zhao *et al*. 2022).

Thermal Stability

Thermal behavior of the bioplastic films was evaluated using thermogravimetric analysis (TGA). The thermogravimetric curves presented in Figure 4(a) reveal that all formulations undergo a

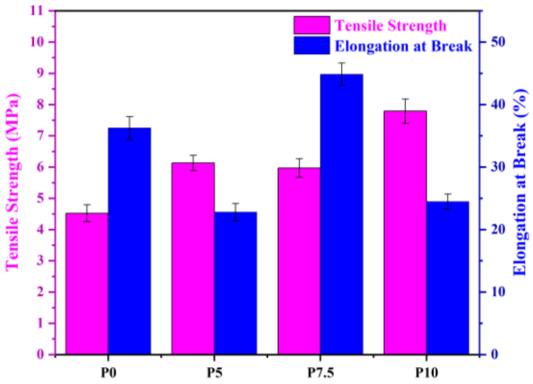

Figure 3. Mechanical performance of the bioplastics.

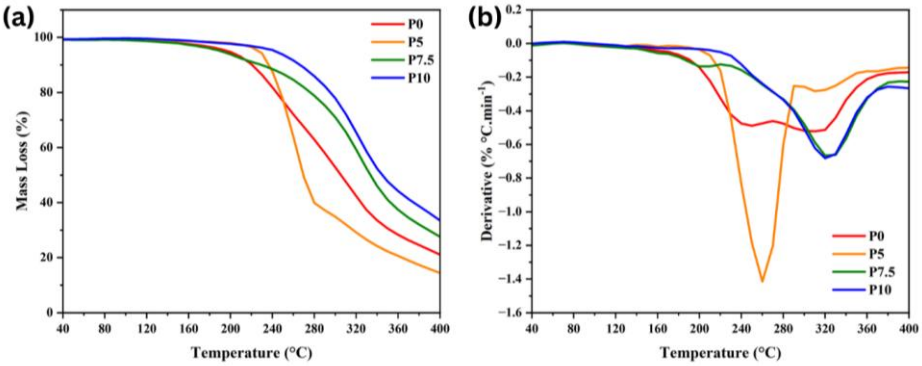

Figure 2. (a) TGA and (b) DTG thermograms

Table 1. Thermal degradation parameters of bioplastic films

Sample	T_{onset} (°C)	T_{max} (°C)	Residue at 400°C (%)
P0	230	290	21.12
P5	240	255	14.44
P7.5	235	320	27.54
P10	260	330	33.47

characteristic stepwise mass-loss process. The initial stage is associated with the evaporation of physically bound water and other volatile components present in the hydrophilic polymer network. This stage is followed by the principal thermal decomposition of the biopolymer constituents, including starch, chitosan, and PVA, which accounts for the largest reduction in sample mass. Such degradation behavior is widely observed in starch-based materials and polymer systems containing PVA (Bracciale *et al.* 2024). The derivative thermogravimetric (DTG) profiles in Figure 4(b) further confirm this behavior, with the maximum degradation temperature shifting toward higher values as PVA content increased, indicating improved thermal resistance. This enhancement is likely associated with stronger hydrogen-bond interactions and improved structural ordering within the films. FTIR analysis suggests enhanced association among starch, PVA, and chitosan chains, while XRD results indicate higher crystallinity in the PVA-rich formulation. These structural characteristics restrict chain mobility and delay thermal decomposition. A summary of the thermal degradation parameters, including T_{onset}, T_{max}, and residual mass at 400°C, is presented in Table 1 to provide a clearer comparison of thermal stability among the samples. Similar improvements in thermal stability have been reported for PVA-based composite films reinforced with MCC or other fillers, where stronger molecular cohesion increases degradation temperature (Hanh *et al.* 2024).

SEM

Figure 5 shows the SEM micrographs illustrating the morphology of the starch-based bioplastic films**.** The control sample (Figure 5(a)) displays a comparatively smooth yet slightly heterogeneous surface, indicating a less uniform film structure. After the addition of PVA, noticeable changes in surface morphology are observed. The P5 film (Figure 5(b)) exhibits a more irregular texture with several visible pores, suggesting that the film network remains relatively loose at this composition. The intermediate formulation P7.5 (Figure 5(c)) presents a more uniform morphology with fewer large pores, indicating improved compatibility among the film components. In contrast, the P10 film (Figure 5(d)) demonstrates the most compact and dense morphology with minimal structural defects, reflecting stronger cohesion within the polymer matrix. This morphological evolution agrees with the density and XRD results, which indicate closer chain packing and greater structural ordering at higher PVA content. Studies on PVA-based composites indicate that improved compatibility between the components promotes the formation of denser microstructures and enhanced mechanical performance (Jannah *et al.* 2021; Wang *et al.* 2024).

Antibacterial Activity

The antibacterial activity of the bioplastic films against *Staphylococcus aureus* and *Escherichia coli* is presented in Figure 6, while the corresponding inhibition zone diameters are summarized in Table 2. The control formulation (P0) exhibited the strongest antibacterial activity, whereas increasing PVA content generally reduced the inhibitory effect of the films. A moderate inhibition zone was still observed for the P5 sample, while the P7.5 and P10 formulations produced noticeably smaller zones of inhibition, with P7.5 showing the lowest antibacterial activity. In all formulations, the inhibition effect was more pronounced against *S. aureus* than against *E. coli*. This difference is often attributed to the cellular

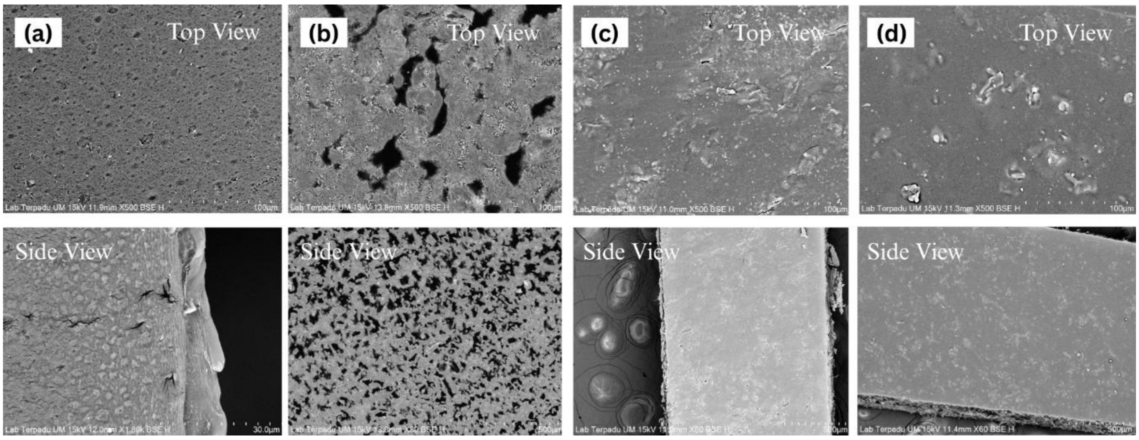

Figure 5. SEM images: (a) P0, (b) P5, (c) P7.5, and (d) P10

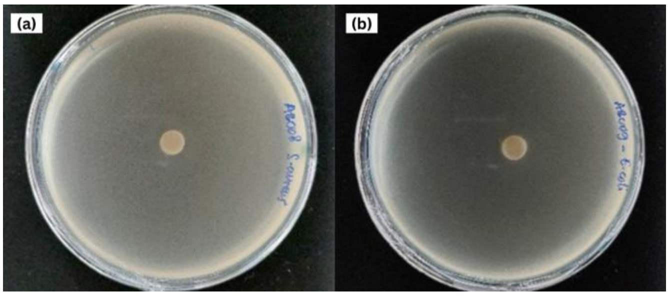

Figure 6. Antibacterial activity against (a) *Staphylococcus aureus* and (b) *Escherichia coli*

Table 2. Antibacterial inhibition zone

Sample	*S. aureus* (mm)	*E. coli* (mm)
P0	12.6	8.8
P5	9.7	8.6
P7.5	4.03	3.43
P10	5.85	5.05

architecture of Gram-negative bacteria, which possess an external membrane layer that acts as an additional protective barrier and restricts the diffusion of antimicrobial substances into the bacterial cell (Venkatesan *et al*. 2022).

The antimicrobial behavior in this system is mainly attributed to the presence of chitosan, whose protonated amino groups are capable of interacting with bacterial cell membranes and disturbing their physiological processes (Ningrum *et al*. 2025). The decline in antibacterial effectiveness with increasing PVA levels may result from structural alterations within the films, as higher PVA fractions enhance hydrogen bonding interactions and lead to more compact polymer chain arrangements, as indicated by FTIR and XRD analyses. These structural characteristics may reduce the accessibility of reactive functional groups from starch and chitosan, such as –OH and –NH groups, that contribute to antibacterial interactions (Putra *et al*. 2025). The more compact morphology observed in SEM images also suggests reduced pore connectivity, which may limit contact between antimicrobial functional groups and bacterial cells. Similar behavior has been reported in polysaccharide-based composite films for active packaging, where increased structural compactness can reduce effective antimicrobial interactions at the material surface (Iqbal *et al*. 2020).

CONCLUSION

This study demonstrates that the performance of starch-based bioplastic films can be effectively tuned by adjusting the PVA content. Increasing PVA concentration promotes stronger intermolecular hydrogen bonding and improved molecular packing, leading to a denser polymer network as confirmed by FTIR, XRD, and SEM analyses. These structural changes enhance tensile strength and thermal stability of the films. However, higher PVA fractions reduce elongation at break, antibacterial activity, and biodegradation rate due to restricted accessibility of functional groups and tighter polymer chain packing. Overall, the results highlight the critical role of intermolecular interactions, crystallinity, and morphology in governing the multifunctional performance of starch-based bioplastics. The findings provide valuable insights for designing biodegradable polymer blends derived from agricultural residues for sustainable active packaging applications.

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