



Sustainable PVA-Chitosan Biocomposite Films: Effects of Chitosan Loading on Material Properties and Biodegradability

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Abstract. Global reliance on non-biodegradable petroleum-based plastics is a significant environmental concern that necessitates the development of eco-friendly alternatives. Poly(vinyl) alcohol (PVA) shows high potential, but its application is limited because of its high moisture sensitivity and intermediate strength. In this study, the effect of crosslinking chitosan (CS) at 5-35% loading on different material properties and biodegradability of poly(vinyl) alcohol-chitosan (PVA-CS) biocomposite films was investigated. FTIR analysis confirmed the successful crosslinking between the two polymers. The integration of CS in the polymer matrix of PVA improved its UV-blocking properties by 25-55%. The degree of swelling and solubility is inversely related to %CS in PVA-CS films. Pure PVA and PVA-CS films showed higher tensile strengths but lower elongation at break compared to LDPE. Thermogravimetry showed 4-stage thermal degradation, with maximum degradation at 280-380°C for the PVA-based films. Significant improvement in the biodegradability of PVA films was observed with the incorporation of CS. Based on the tested %CS loading, a minimum of 15% CS is required to see significant improvement in the biodegradation rate, UV-blocking, and mechanical properties of the PVA-CS films. These findings have direct practical implications for sustainable engineering, showing the potential of PVA-CS biocomposites as a biodegradable alternative to traditional plastics.

Keywords: biodegradable polymer, bioplastic film, polymer crosslinking, solution casting, sustainable material, UV-blocking

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1. Introduction

Plastic pollution is among the most pressing environmental issues that global stakeholders face. With annual production reaching approximately 400 million tons, the environmental persistence of these non-biodegradable materials renders traditional waste disposal and management methods infeasible [1]. This necessitates a shift toward more sustainable materials, including biodegradable plastics and biocomposite films, which have been the subject of several studies in recent years [2]. While research on biocomposites has surged, the widespread adoption of these materials is often hindered by the associated trade-offs in terms of performance and cost.

Poly(vinyl) alcohol (PVA) has been studied as a promising alternative to traditional synthetic plastics due to its biodegradability and mechanical properties [3, 4]. However, its high production cost and functional limitations compared with its synthetic, non-biodegradable counterparts remain significant barriers to industrial scalability [5]. To address this, filler materials are commonly incorporated into the PVA polymer matrix to create a composite material that is cheaper and has superior properties [5, 6].

Several studies have explored preparing composites by combining PVA with chitosan (CS), a chitin derivative. Chitosan has numerous favorable properties, including non-toxicity, biodegradability, high polymer crystallinity, strong intermolecular forces, and biocompatibility, which enable its use in a wide range of fields, such as pharmaceuticals, food, agriculture, and cosmetics [7]. In the context of the circular economy, chitosan can be sourced from industrial food waste, such as fish scales, shrimp and crab shells, squid pens, and fungi [8]. When combined with other polymers to form bioplastic and biocomposite materials, chitosan can significantly improve the biodegradability of these polymers, making them suitable for single-use plastic applications, such as food packaging films [9].

Two common methods for producing PVA-CS biocomposite films are blending and crosslinking [10]. The key difference is that blending involves only physical mixing of two miscible polymers [11, 12], whereas crosslinking adds crosslinkers to chemically bond the two polymers [13, 14]. PVA-CS blended films exhibit antimicrobial, antioxidant, and UV-barrier properties, along with moderate mechanical properties [15-17]. However, the properties of such blends can vary significantly depending on the miscibility of the polymer components. For example, PVA-CS films produced by blending often lack flexibility, have low thermal stability, and are highly water-soluble [18, 19]. These properties can be significantly enhanced by crosslinking the two polymers [20].

Glutaraldehyde is a common crosslinking agent for PVA and chitosan and has been shown to improve the mechanical strength and water resistance of the resulting composite [21-23]. Furthermore, when used as a crosslinking agent, glutaraldehyde can alter chitosan's hydrophilicity and improve the sensitivity to acidic solutions and the barrier properties of CS films, among other effects [21, 24]. These molecular reinforcements ensure that sustainable materials, such as PVA-CS biocomposites, can withstand real-world applications, logistics, and varying storage conditions.

Although some studies have already explored the preparation and characterization of PVA-CS films, the literature remains limited on the effect of CS content on the physicochemical and mechanical properties of PVA-CS biocomposites, with most studies conducted on polymer blends rather than crosslinked biocomposites [25]. For crosslinked PVA-CS, CS loading has been varied, but only at low loadings below 5% [16] or at very high loadings greater than 25%, up to more than 50%, which may suggest a role primarily as a reinforcer [18].

In this study, biocomposite films were synthesized with varying CS amounts (5%, 15%, 25%, and 35%) relative to the PVA mass via the solution-casting method, using glutaraldehyde as a crosslinker. These loading percentages were selected because they are intermediate relative to what is typically used

in the literature, and some of these CS loadings remain unexplored, especially for crosslinked films. Glutaraldehyde was chosen as a crosslinker because it is highly reactive and can form stable covalent bonds more quickly, even at lower heating temperatures. Other crosslinkers, such as citric acid, usually require higher processing temperatures, which can degrade sensitive biopolymers.

The physicochemical and mechanical properties of the synthesized films were investigated and compared with those of commercially available plastics, including LDPE and DOST ITDI ETV-13-005 Oxo-biodegradable HD bags. LDPE is a flexible, low-density material composed of branched chains of ethylene monomers, exhibiting excellent moldability and barrier properties [26]. It was chosen as a positive control due to its widespread use in industrial applications. The DOST Oxo-biodegradable bag was chosen as a positive control for biodegradability testing because the product was marketed as wholly biodegradable and received an Environmental Technology Verification (ETV) certification from the Department of Science and Technology - Philippines.

2. Methods

2.1. Materials

Technical-grade PVA BP-24 was obtained from Dalkem Corporation, Philippines. Commercial chitosan was obtained from Xi'an Pincredit Bio-Tech Co., Ltd., China, while glutaraldehyde (GA) was obtained from Isamu Chemical International Corporation, Philippines.

2.2. Synthesis of Pure PVA and PVA-CS films

Pure PVA and PVA-CS biocomposite films were prepared by solution casting and solvent evaporation, following the protocols of Pugar et al. (2024) and Costa et al. (2009) with some modifications [13, 27]. A predetermined mass of PVA powder was dissolved in 300 mL of distilled water at room temperature and heated to 80 °C for 1 hr until complete dissolution. In a separate beaker, a CS solution was prepared by dissolving a specified amount of CS in 100 mL of 2% acetic acid. The CS solution was then mixed with the PVA solution, followed by the addition of 1 mL of 1% glutaraldehyde (GA) solution.

Table 1. Polymeric Film Composition (% mass polymer/total mass)

Sample	%PVA	%CS
PVA	100	0
PVA-5%CS	95	5
PVA-15%CS	85	15
PVA-25%CS	75	25
PVA-35%CS	65	35

Although studies have shown that higher GA crosslinking concentrations (2.5-7.5%) [28] yield better mechanical properties, the use of 1% GA ensures that all of the GA is completely consumed in the reaction. Although no direct measurement of residual GA was performed in this study, the low standard deviation in our tests suggests that the crosslinking was uniform and that the agent was efficiently integrated into the polymer matrix. The ratio between the total volume of solvent and the total mass of polymer mix was maintained at 30:1 for all samples. The solution was stirred for 1 hr with minimal heating, then degassed by sonication at 60°C for 30 min [29]. The solution was then cast into 10 cm x 10 cm x 0.5 cm silicone molds and dried at 60°C for 24 hrs. The produced films, whose composition is presented in Table 1, were stored in a desiccator for further testing.

To better understand the effects of chitosan as a filler, intermediate concentrations at 10% intervals between the upper (35%) and lower limits (5%) were examined. These intermediate concentrations were deemed necessary to provide insight into the specific effects of using chitosan as a reinforcer to improve the physicochemical and mechanical properties of PVA/CS composites. We did not include higher CS loadings, as these are well explored in the literature, and if CS exceeds 50%, it will serve as the main polymer matrix instead of PVA.

2.3. Fourier Transform-infrared (FTIR) Analysis

Fourier Transform-Infrared (FTIR) Spectroscopy (Agilent Cary 630 FTIR ATR) was used to confirm successful crosslinking and synthesis of the PVA-CS films. IR spectra of the samples were obtained by measuring the % transmittance from 600 to 4000 cm⁻¹.

2.4. Optical Properties

The optical properties of the films were measured using a UV-visible spectrophotometer (UV-1800, Shimadzu) to determine the average light transmission in the UV range (250-400 nm) and the visible range (400—780 nm). A 1 cm x 4 cm film was placed in the test cell, and an empty cell served as the reference. The film's opacity was calculated using the equation:

$$\text{Opacity} = \frac{\text{Abs}_{600}}{x} \quad (1)$$

where Abs₆₀₀ and x are the absorbance at 600 nm and film thickness (mm), respectively [30, 31].

The average transmittance (T_{Ave}) of the film was calculated using the equation:

$$T_{\text{Ave}} = \frac{\int_{\lambda_1}^{\lambda_2} T(\lambda) d\lambda}{\lambda_2 - \lambda_1} \quad (2)$$

where T(λ) is the transmittance at any wavelength (λ). λ₁ and λ₂ represent the lower and upper limits of the wavelength, respectively. The light transmittance of the film samples at 350 nm and 500 nm was also measured and compared across all films [32-34].

2.5. Degree of Swelling and Water Solubility Test

The effect of pH on the swelling of PVA-CS films was investigated using the method of Miriam et al. (2024). A 1 cm x 1 cm dry sample of the PVA and PVA-CS films was immersed in 10 mL of buffer solutions at pH 4, 7, and 10 at room temperature. The samples were weighed at 30, 60, 90, and 120 min, and mass changes were recorded. Before weighing, the samples were blot-dried to remove excess media from the surface [35]. The degree of swelling was calculated using the equation:

$$\text{degree of swelling (SD)} = \frac{M_s - M_d}{M_d} \quad (3)$$

where M_d and M_s are the weights of the dry and swollen samples measured at each time point [22, 36].

The equilibrium swelling degree (ESD) of the films was also determined gravimetrically by allowing a 1 cm x 2 cm sample to swell for 24 hours in 50 mL of water. The excess water was removed with filter paper, and the mass of the swollen films was measured. ESD for each film was calculated using the equation:

$$\text{Equilibrium Swelling Degree (ESD)} = \frac{M_s' - M_d}{M_d} \quad (4)$$

where M_s' is the weight of the samples swollen for 24h [37, 38].

The water solubility of the samples was determined by drying the swollen samples for 24 hrs at 60 °C, and it was computed using the equation:

$$\text{Solubility (S)} = \frac{M_d - M_{d,s}}{M_d} \times 100 \quad (5)$$

where M_{d,s} is the dry weight of the swollen films [19, 39].

2.6. Mechanical Property Testing

The mechanical properties of the synthesized films were evaluated using a Universal Testing Machine (Shimadzu AGS-V). Samples were conditioned at 25°C and 50% relative humidity for 40 hours. After conditioning, the samples were cut into 30 mm x 110 mm strips (triplicate). The following equations were used to determine the tensile strength and elongation at break of the biocomposite films:

$$\text{Tensile strength (MPa)} = \frac{F_M}{A_0} \quad (6)$$

$$\text{Elongation at break (\%)} = \frac{L_E - L_0}{L_0} \quad (7)$$

where F_M is the maximum stress, A₀ is the film's original cross-sectional area, L is the film's extension at the moment of rupture, and L₀ is the film's initial length. The Young's Modulus was calculated as the

slope of the first linear region of the stress-strain curve [23], while the yield strength was determined using the 0.2% offset strain method [40].

2.7. Thermal Stability

The thermal stability of the synthesized films was tested using a thermogravimetric analyzer (NETZSCH STA 2500 Regulus). A 1-2 mg film sample was placed in a crucible and then heated from 25 °C to 700 °C at a ramping rate of 10 K/min [41].

2.8. Biodegradability

The biodegradability of the biocomposite films was tested according to ASTM D5988, as used by Abdullah and Dong (2019), with slight modifications. 2 L plastic containers were filled with sieved loam garden soil obtained from a local online store. Biocomposite film samples were cut into 3 cm x 3 cm pieces and weighed to record the initial weight (W_0). Samples were buried at a depth of 4 cm below the surface. This depth was chosen to simulate topsoil conditions, which are considered the most biologically active zone. After 30 days, the samples were extracted from the containers, washed with distilled water, and dried in a drying oven overnight. A 30-day incubation was chosen to allow the establishment of microbial colonies and to determine whether the films are rapidly biodegradable. Once dried, the weights of the samples were recorded (W_T). The following equation was used to determine the biodegradation rate [5]:

$$\text{Biodegradation rate (\%)} = \frac{W_0 - W_T}{W_0} \times 100 \quad (8)$$

Furthermore, a comparative analysis between the synthesized biocomposites and DOST ITDI ETV-13-005 Oxo-biodegradable HD bags, used as a positive control, was conducted to evaluate the properties of the biocomposites against other available synthetic commercial plastics.

2.9. Statistical Analysis

All analyses were performed in triplicate. The low standard deviation observed across characterization tests confirmed that the film fabrication process is stable and repeatable, supporting the use of triplicate measurements. The data were subjected to a one-way analysis of variance (ANOVA) with Tukey HSD post hoc test to evaluate significant differences between the samples at a significance level of $p \leq 0.05$. In addition, a comparative analysis was conducted on the reinforced biocomposite films, DOST ITDI ETV-13-005 Oxo-biodegradable HD bags, and LDPE.

3. Results and Discussion

3.1. FTIR Analysis

The films underwent FTIR analysis to confirm successful crosslinking of PVA and CS in the synthesized composite films. The FTIR spectra of chitosan, pure PVA, and PVA-CS films are shown in Figure 1.

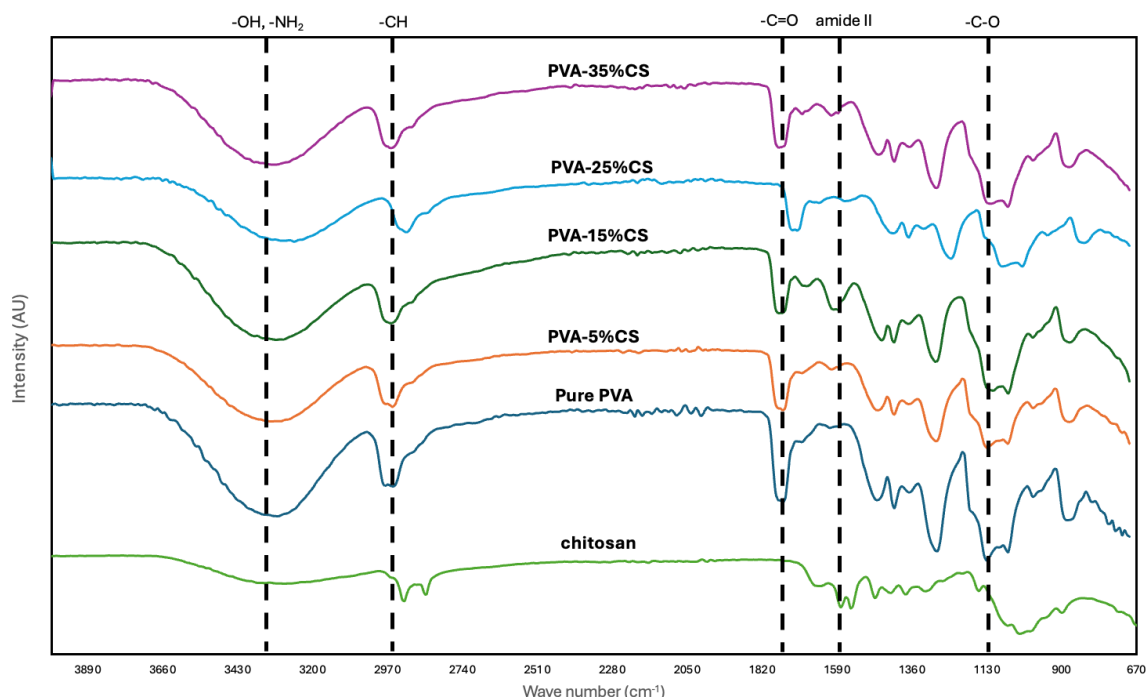


Figure 1. FTIR spectra of chitosan, pure PVA film, PVA-CS biocomposite films, and LDPE.

The chitosan IR spectrum showed distinct peaks around 3300 cm^{-1} , 2950 cm^{-1} , and 1590 cm^{-1} . The broad peak at $3200\text{--}3600\text{ cm}^{-1}$ corresponds to -OH , with overlaps of the -NH_2 groups and intermolecular hydrogen bonds, while the peak at 2950 cm^{-1} is attributed to CH stretching [25, 42]. The peak around $1549\text{--}1584\text{ cm}^{-1}$ corresponds to amide II groups resulting from the N-deacetylation of chitin to form chitosan [43]. The absence of a C=O stretch in chitosan suggests a high degree of deacetylation [44]. In PVA, distinct peaks for -OH ($\sim 3300\text{ cm}^{-1}$), -CH ($\sim 2930\text{--}2980\text{ cm}^{-1}$), -C=O ($\sim 1720\text{--}1780\text{ cm}^{-1}$), and -C-O stretching ($\sim 1290\text{--}1330\text{ cm}^{-1}$) were present in the IR spectra [42, 45]. These peaks were also present in all PVA-CS composites.

The polymeric network in PVA-CS composites forms through the interaction of the aldehyde group of glutaraldehyde and the amine group of chitosan, resulting in imine bonds (C=N). Similarly, -OH groups in PVA can also interact with the aldehyde groups of glutaraldehyde. The crosslinking reaction will proceed until all the -OH groups of PVA and the amino groups of chitosan have reacted with the aldehyde group of glutaraldehyde via nucleophilic addition [46]. With this, we can confirm successful crosslinking by observing the presence of an imine group (C=N) and an acetal group (C-O) in the IR spectra of the PVA-CS films.

Chemical crosslinking of PVA and chitosan was confirmed by the presence of a peak around $1640\text{--}1690\text{ cm}^{-1}$, corresponding to C=N stretching, in all PVA-CS films. Small, sharp peaks around $1530\text{--}1540\text{ cm}^{-1}$ can be attributed to -NH_2 stretching. The sharp peak around 1250 cm^{-1} is attributed to the C-O bond of PVA. On the other hand, the broad peak between 1030 and 1080 cm^{-1} is attributed to the C-O bonds of chitosan and to the C-O bonds formed between glutaraldehyde and polyvinyl alcohol. The peaks present in the FTIR spectra are similar to those reported in previous studies [47–49].

3.2. Optical Properties

UV-vis spectroscopy was performed on the different films to assess their light-barrier properties by measuring their light transmittance (Figure 2). Initial visual observations show that the color of the films shifts from colorless to a transparent yellow as the chitosan loading increases. This is due to the natural yellowish color of chitosan when dissolved in solution.

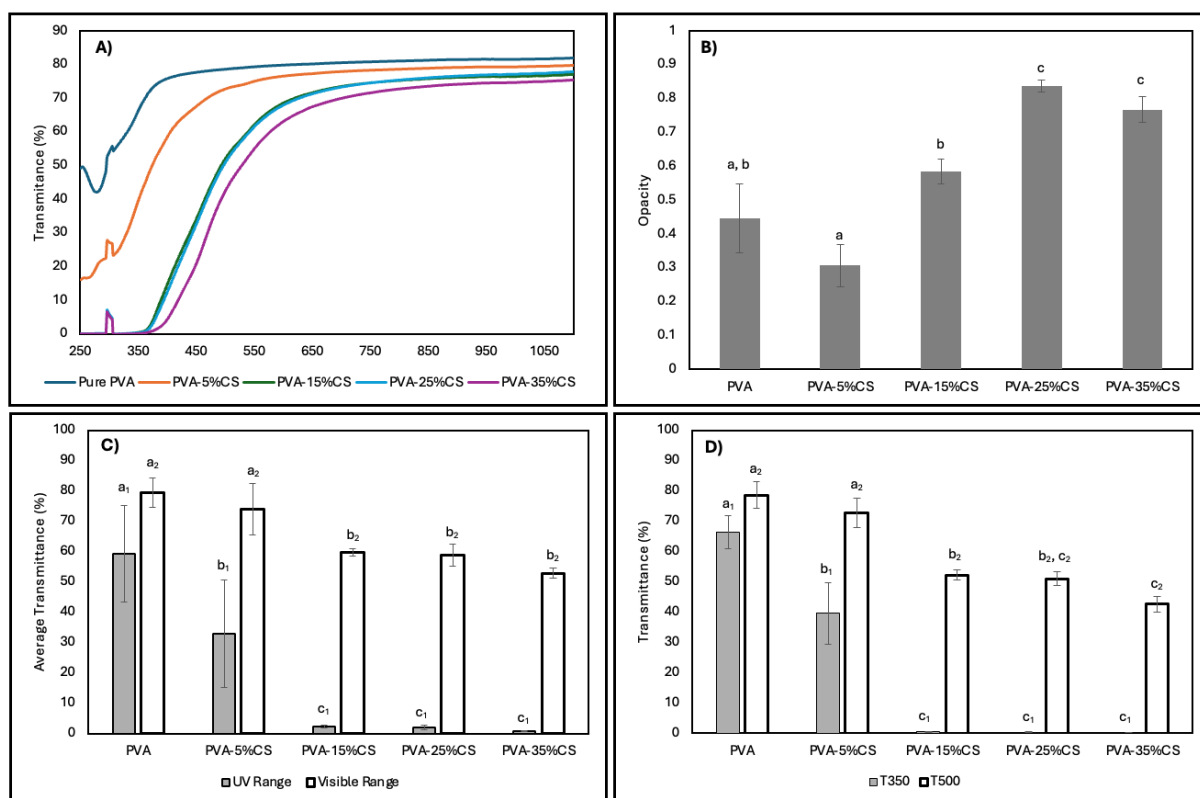


Figure 2. A) UV-vis transmittance, B) Opacity, C) Average transmission in the UV and visible range, and D) T₃₅₀ and T₅₀₀ of PVA and PVA-CS films. Different letters above the bars represent a significant difference between the treatments for the same wavelength range based on ANOVA ($p < 0.05$) and Tukey's HSD. Numbers beside the letters indicate data being compared (1: UV-region, 2: Visible region).

Results show that pure PVA has higher transmittance across all wavelengths than the PVA-CS composites (Figure 2A). Incorporating CS into the PVA matrix significantly reduces transmittance. PVA-CS films with higher CS content exhibit better UV-blocking properties than pure PVA and PVA-5% CS. This is confirmed by the average transmittance in the UV region (Figure 2C), where pure PVA has a significantly higher transmittance (59%), compared to PVA-15%CS, PVA-25%CS, and PVA-35%CS, which have a mean transmittance of only 1-2%. The apparent color imparted by chitosan to the films, which is more obvious at higher CS loading, results in a decrease in transmittance in the visible range.

The same trends were observed for the T₃₅₀ and T₅₀₀, which also translate to lower computed opacity values (Figure 2B). Higher opacity values indicate that the films are less transparent, which, in this case, could indicate a homogeneous dispersion of the components of the film [50]. This increase in the film's opacity and improved UV-blocking property for the PVA-CS films can be advantageous for applications such as UV-protective coatings for windows and food packaging, where UV exposure can promote food degradation through UV-induced lipid oxidation reactions [34].

3.3. Swelling Behavior and Solubility

PVA is a well-known polymer with high water solubility, which is why it is used to improve the solubility of lipophilic drugs [51] and to develop hydrogels whose polymer network can accommodate water molecules [52]. The pH-dependent swelling experiments showed that, at all tested pH levels (4, 7, and 8), PVA-5%CS exhibited the highest swelling degree, even higher than that of the pure PVA film.

This may be due to the strong tendency of the amino groups of CS to form hydrogen bonds with water in the swelling medium [53].

Results also showed an inverse relationship between CS content and the degree of swelling across all pH conditions. This is attributed to the reduced ability of the films to form hydrogen bonds with water molecules after crosslinking CS and PVA via acetalization and Schiff base formation [36]. Our results are consistent with previous studies that have observed increased resistance to swelling at higher CS concentrations under more acidic conditions [35]. After the rapid initial swelling at 30 min, a minimal increase in swelling was observed at the other time points for most films.

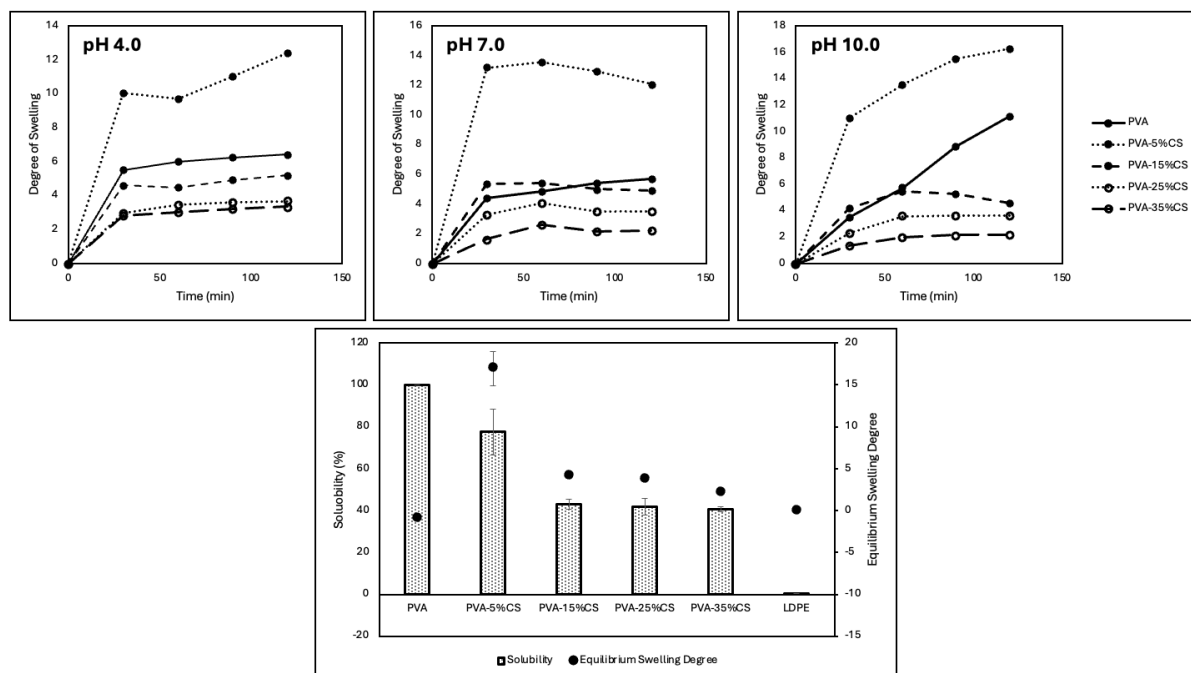


Figure 3. pH-dependent swelling, equilibrium swelling degree, and solubility of PVA, PVA-CS composite films, and LDPE.

Equilibrium swelling degree (ESD) and water solubility (WS) were determined for PVA, PVA-CS films, and LDPE by immersing the films in distilled water at room temperature for 24 hours. The -OH and -NH₂ groups in PVA and PVA-CS films can form hydrogen bonds with water [54], allowing the films to absorb water and swell. The highest ESD was observed for PVA-5%CS, consistent with our pH-dependent swelling experiments. Although the pure PVA film is expected to take up the most water during swelling, it completely dissolved during the 24 h immersion, leaving only small amounts of residue when the solvent was removed. When immersed in solution, PVA readily absorbs water because of its hydrophilic properties, forming hydrogen bonds with water. Eventually, PVA chains separate, resulting in dissolution of the polymer.

Both the ESD and the water solubility of the PVA-CS films decreased with increasing %CS loading. The low swelling degree observed at higher %CS confirms extensive crosslinking between PVA and CS [55]. Negligible swelling and dissolution were observed for LDPE after 24 hrs of exposure, which is expected given the hydrophobic properties of LDPE [56]. The decrease in solubility of the PVA-CS films indicates the potential of this biocomposite for use in outdoor or humid conditions without immediate failure. Films with a lower %CS can be used for water-soluble packaging. Its low solubility and high swelling properties also allow for its application in the sustainable release of nutrients in agriculture.

3.4. Mechanical Properties

It is important to study the mechanical properties of films because parameters such as elongation at break and tensile strength can provide information about their durability and potential applications. Results for tensile strength, yield strength, Young's modulus, and elongation at break for the different films are summarized in Figure 4.

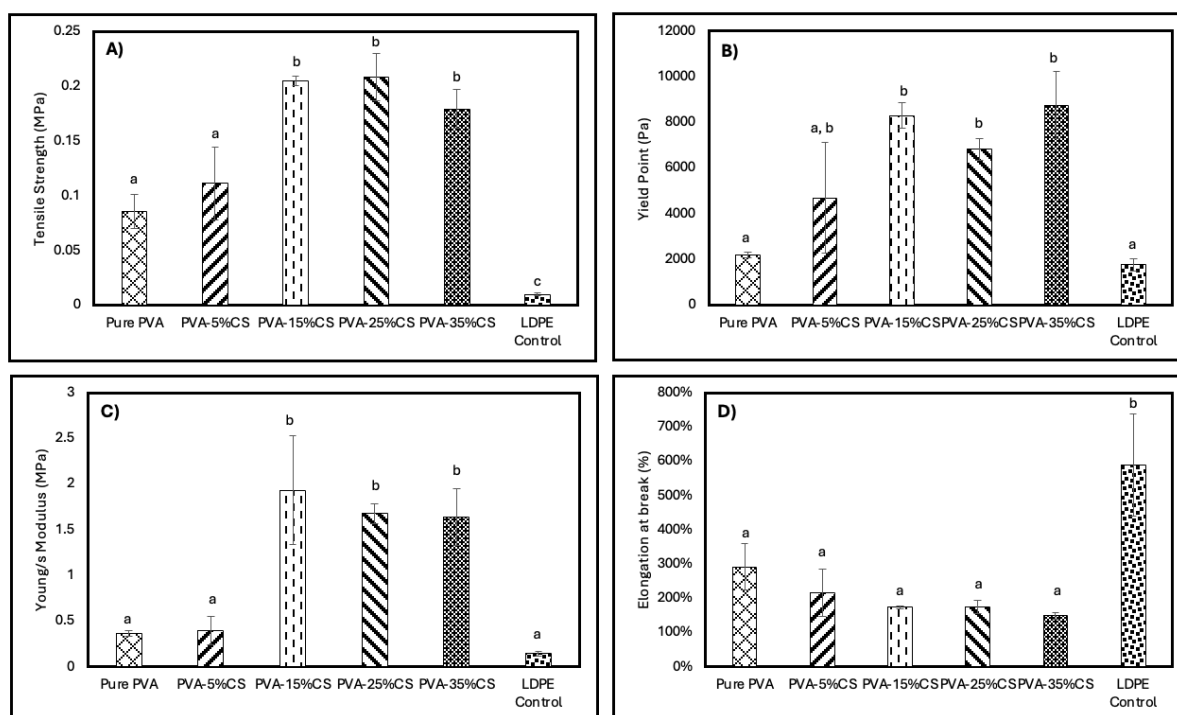


Figure 4. A) Tensile strength, B) yield strength, C) Young's modulus, and D) elongation at break of the synthesized biocomposite films and LDPE. Different letters above the bars represent a significant difference between the treatments for the same wavelength range based on ANOVA ($p < 0.05$) and Tukey's HSD.

Pure PVA and all PVA-CS composite films showed significantly higher tensile strength than LDPE. Conversely, elongation at break was highest for LDPE. These results indicate that the LDPE control has greater flexibility and durability than pure PVA and PVA-CS films. A 5% CS loading did not significantly improve tensile strength, whereas films with 15-35% CS showed more than a two-fold increase in tensile strength.

Similar trends were observed for Young's modulus and yield strength. No improvement in elongation at break was observed with crosslinking CS to PVA. Previous studies have shown that increasing chitosan concentration in composite films may increase the stiffness and strength of the films, resulting in low elongation at break values [4]. Furthermore, the high tensile strength and Young's modulus for films with higher CS content can be attributed to increased hydrogen-bonding interactions. This same interaction can also limit the movement of molecular segments, resulting in the apparent decreasing trend in the elongation at break of the films [57].

Integrating CS into the PVA matrix shifted the material from a soft, ductile polymer to a high-strength biocomposite. The higher yield point at 15-35% CS enables the design of thinner films that can withstand stacking and heavy loads without deforming. Although its rigidity and low elongation at break limit its use as bags, PVA-CS can be used for other high-demand applications such as rigid food trays, protective lids, and secondary packaging.

3.5. Thermogravimetry

Thermogravimetric analysis was performed on the films to study changes in material composition and to determine the thermal stability of the films. The TGA and DTG curves for the different films, including the LDPE control, are shown in Figure 5. The onset temperatures of the PVA-CS films were compared with those of the LDPE films. Furthermore, the significant weight-loss stages of the pure PVA, PVA-CS, and LDPE films were determined and analyzed.

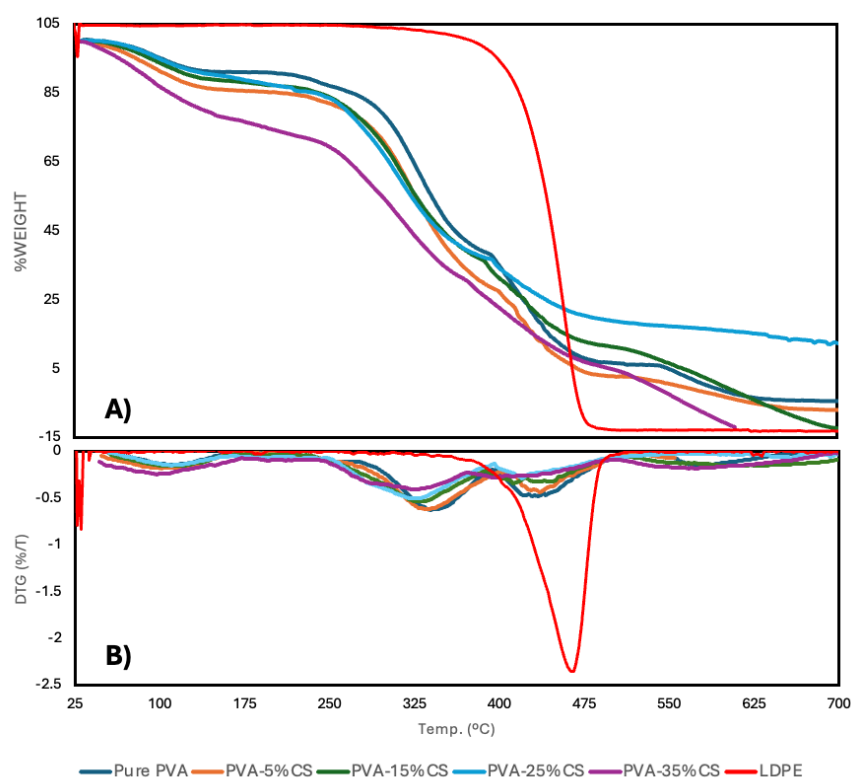


Figure 5. A) TGA and B) DTG curves of pure PVA, PVA-CS composite, and LDPE films.

Peaks in the DTG curves indicate the temperatures at which the maximum slope of each degradation stage occurs [58]. The DTG graphs for pure PVA and PVA-CS films show that the samples undergo a 4-stage thermal decomposition process. This corresponds to four weight-loss stages observed in the thermograms of all the PVA-based films. The first weight-loss stage occurs between 80°C and 118°C and is attributed to the loss of moisture (H₂O) and vaporization of solvent residues, particularly acetic acid used to increase the solubility of chitosan [59]. This stage accounts for 6-18% of the weight loss in all tested films. The -OH groups of both chitosan and PVA, together with the -NH₂ groups of chitosan, enable strong interactions with water, which can be removed once the appropriate temperature is reached.

The second stage of weight loss, which accounts for 40-55% of mass loss in the samples and occurs between 250°C and 380°C, can be attributed to the degradation of PVA-chitosan and the formation of PVA decomposition products such as aldehydes, alkenes, and polyenes [58, 60]. This stage also marks the onset of degradation of the exposed side chains, in addition to the polymeric chain breakdown of both PVA and chitosan [49].

The third stage of weight loss, occurring between 380 °C and 475 °C, is associated with the total degradation of the biocomposite and the degradation of PVA decomposition byproducts [60]. Because the chitosan used was not further purified and was used as is, calcium hydroxide produced during the deacetylation of chitin can undergo thermal dehydroxylation at this temperature, followed by

decarboxylation of the calcite and carbonate phases formed [43]. Oxidative decomposition of the carbon residues also occurs in this stage, before total decomposition to ash at temperatures above 500°C [49].

For the LDPE, only one weight-loss stage was observed near 420°C, attributed to the decomposition of carbonated chains [61]. The LDPE was also stable up to around 440°C. The LDPE showed the highest onset temperature among all the samples, indicating superior thermal stability compared to the PVA-based films. CS loading in the PVA-CS biocomposites shifts the onset temperature to a slightly lower value relative to the pure PVA film, indicating reduced thermal stability. Still, the onset temperature observed for all PVA-based films is around 300°C, allowing for a variety of applications not involving very high-temperature exposure, including food packaging, plastic mulch in agriculture, and medicine. However, the lack of a test for residual glutaraldehyde in the study limits the application of the glutaraldehyde-crosslinked PVA-CS biocomposite films to non-food contact and non-medical applications.

3.6. Biodegradability

The biodegradability rate (BR) of the PVA-CS films was measured using a 30-day soil burial test, with percent mass loss recorded after the experiment. The BR of the PVA-CS films was then compared to that of pure PVA films and DOST ITDI ETV-13-005 Oxo-biodegradable HD bags (Figure 6).

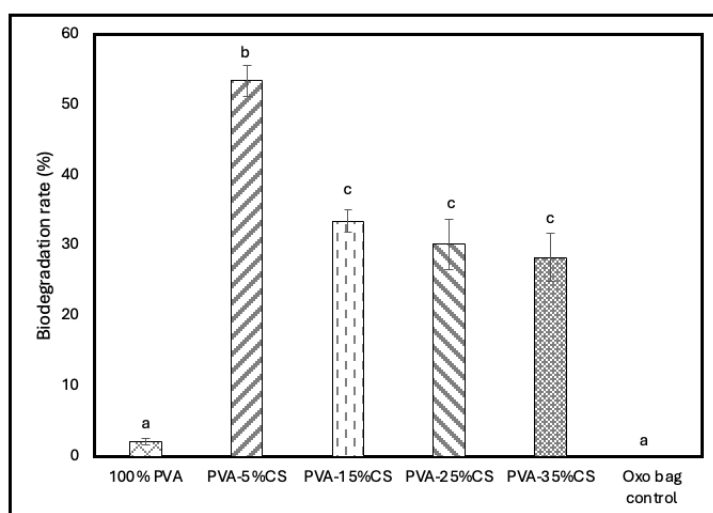


Figure 6. Biodegradability of pure PVA, PVA-CS, and Oxo bag control. Different letters above the bars represent a significant difference between the treatments for the same wavelength range based on ANOVA ($p < 0.05$) and Tukey's HSD.

All PVA-CS films had a significantly higher BR than the pure PVA films and Oxo-biodegradable HD bags. The 5% CS films also showed a significantly higher BR than the pure PVA films. The stark difference between the two films may be attributed to chitosan's inherently high biodegradability [8, 62]. Among PVA-CS films, the BR decreased as the PVA concentration increased. However, the 15% CS, 25% CS, and 35% CS showed similar BR, whereas the 5% CS showed an exceptionally high BR.

The similarity in biodegradability profiles of films with high concentrations of chitosan may be due to their similar hydrolyzable properties. The high BR of PVA-5%CS may be connected to its higher water uptake. When water molecules enter the polymer matrix, they can disrupt the H-bonding between the polymer chains, contributing to increased biodegradability of the composite films [57]. Furthermore, moisture in soil allows for the initial stages of hydrolysis in biodegradable polymers, resulting in breakdown of the polymer chain [63]. This makes the films more susceptible to microbiological degradation. Azahari et al. (2011) also reported that the biodegradability of PVA-based films may be

affected by enzymatic action, and as a result, the trend of biodegradability is expected not to follow a simple linear relationship with filler concentration [64].

3.7. Industrial prospects and safety considerations

The produced PVA-CS biocomposite films showed a competitive property profile when benchmarked against other bioplastic films and their non-biodegradable counterparts (Table 2). Crosslinked PVA-CS films exhibited significantly improved biodegradability compared to pure PVA. Although the tensile strength and maximum degradation temperature remain lower than those of synthetic benchmarks such as LDPE and polypropylene, the films still demonstrated superior elongation at break compared to many starch-based bioplastic films. The balance of thermal stability and flexibility suggests that crosslinked PVA-CS biocomposites are viable, sustainable alternatives to non-biodegradable packaging materials.

Table 2. Comparison of properties of selected bioplastic films and non-biodegradable counterparts.

Type of Polymer/Material	Water solubility	Tensile strength (MPa)	Elongation at break (%)	Maximum degradation temperature (°C)	Biodegradability	References
Pure PVA	100%	0.08 - 0.1	300%	325	2% (after 30 days)	In this study
Crosslinked PVA-CS	40-85%	0.08 - 0.23	180 - 230%	320 - 325	25-58% (after 30 days)	
Dialdehyde starch-modified starch	3-10%	1.63 - 2.06	0.85 - 1.1	295 - 297	33 - 47% (after 5 days)	[65]
Silica-modified starch	8-22%	0.53 - 0.75	0.16 - 0.28	246 - 270	63 - 78% (after 5 days)	[66]
Taro starch-bentonite	No data	1 - 2.1	9 - 12.5	No data	19 - 31% (after 10 days)	
Starch-chitosan	No data	7.7 ± 0.45	82 - 238	315 - 321	42 - 96% (after 24 hr)	[67]
Pectin-chitosan	No data	0.4 - 0.6	235 ± 3.5	290 - 350	78 - 96% (after 24 hr)	
Carboxymethyl Cellulose	No data	4.2 - 6.9	14.8 - 16.2	290 - 300	100% (after 7 days)	[68]
ZnO-Carboxymethyl Cellulose	No data	5.2 - 16.2	5 - 12	290 - 300	70 - 82% (after 7 days)	
Carboxylated Cellulose nanofiber	12.2	44.4 - 114.2	7.2 - 8.9	256.5 - 276	No data	[69]
Cellulose from <i>Cladophora sp.</i>	28.88 - 51.91	1.18 - 9.33	3.78 - 38.73	290 - 340	21.25 - 62.54% (after 5 days)	[70]
Protein-pectin	18.72	No data	264.87	200	100% (after 15 days)	[71]
LDPE	-	8 - 31	100 - 965	T _m = 105 - 115	Non-biodegradable	[72]
Polypropylene	-	31 - 43	500 - 650	T _m = 200 - 300 [73]		
Polyethylene terephthalate	-	48 - 270	30 - 3,000	T _m = 160 - 180 [73]		
Polyamide	-	41 - 165	300 - 400	T _m = 180 - 220		

Despite these promising properties, industrial adoption is hindered by the limitations of traditional crosslinkers. Structural reinforcement in this study was achieved using glutaraldehyde, a crosslinker known for significant toxicity and leaching risks. This compromises the status of these biocomposites as green materials and food-safe. Although only 1% GA was used – substantially lower than the commonly used 3-5% range – measurement of residual GA remains imperative to ensure that no toxic GA remains, especially if it is to be used for food and drug applications.

4. Conclusion

This study successfully engineered high-performance crosslinked poly(vinyl) alcohol-chitosan biocomposite films, establishing an optimal range of 15% to 35% to maximize mechanical integrity and UV-shielding efficacy. FTIR analysis confirmed that glutaraldehyde-mediated crosslinking of PVA and CS is the primary driver of improved hydrothermal stability, effectively tailoring the swelling degree and solubility for wet applications. 15-35% CS loading in the composite films significantly improved the mechanical properties of the PVA-CS films. Thermal degradation of the biocomposites is characterized by a 4-stage decomposition process. The biodegradation rate of the PVA-CS films is closely associated with the films' moisture uptake. These properties position these films as superior alternatives to LDPE. However, industrial adoption remains dependent on rigorous safety validation. The findings underscore the immediate viability of PVA-CS biocomposites for medical and food-contact applications. To bridge the gap toward food-grade packaging, future studies must prioritize residual glutaraldehyde quantification to ensure toxicological safety. This can be further extended by including water vapor and oxygen transmission rate measurements and by exploring the use of bio-based, non-toxic crosslinking alternatives. This will be important in fully realizing the potential of these green materials in the global circular economy.

Declaration of AI and AI assisted technologies in the writing process

During the preparation of this work, the author(s) used Grammarly to check spelling and grammar, including punctuation, in the main text. After using this tool/service, the author(s) reviewed and edited the content as needed and take(s) full responsibility for the content of the publication.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could appear to influence the work reported in this paper.

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