

Copper-embedded polyvinyl alcohol (PVOH)-based antimicrobial and biodegradable coating for food packaging applications

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ABSTRACT: The food packaging industry increasingly seeks multifunctional, bio-based materials that ensure product safety while supporting strict municipal environmental sustainability goals. This research addresses functional limitations in compostable packaging by developing a polyvinyl alcohol (PVOH)-based antimicrobial and biodegradable coating through precise incorporation of active copper ions.

Copper ions were incorporated into a 20% solids-content PVOH matrix at target concentrations of 100, 500, and 1,000 ppm, then applied to paper and polylactic acid (PLA) bioplastic substrates at a target coating weight of 6 g/m². Antimicrobial testing against *Escherichia coli* and *Salmonella* spp. using a bacterial overlay agar method demonstrated robust, localized microbial inhibition zones that increased with copper concentration. Biodegradability and industrial compostability were assessed under controlled conditions over 49 days in accordance with ASTM D5338 and ASTM D6400. Uncoated butcher paper controls exhibited 71.90% biodegradation, whereas copper-embedded PVOH-coated paper achieved 83.59%, conclusively demonstrating that integrated metallic ions did not hinder or delay natural macromolecular decomposition. Scanning electron microscopy and elemental analysis verified uniform coating distribution and stable copper dispersion across all treated surfaces. Differential scanning calorimetry revealed minor, predictable modifications to polymer crystallization kinetics without altering macrostructural film integrity. Barrier evaluations (ASTM F1249 and ASTM D3985) confirmed a substantial, highly significant reduction in oxygen transmission rates, whereas the reduction in water vapor transmission rates was not statistically significant due to PVOH's inherently hydrophilic nature. These findings demonstrate that copper-embedded PVOH coatings provide a highly effective, active antimicrobial defense system while remaining completely compatible with commercial, sustainable industrial composting loops.

Application: This study demonstrates the potential of copper-embedded PVOH coatings as a sustainable approach for developing active food packaging materials. These findings provide paper, bioplastic, and bio-based packaging converters with a viable method to design functional barrier coatings that deliver robust antimicrobial defenses without disrupting industrial composting and degradation workflows.

The food-packaging industry faces a dual challenge: ensuring product safety while advancing environmental sustainability. Cross-contamination by pathogenic microorganisms remains a major driver of food-borne illness throughout the supply chain, increasing demand for active, antimicrobial packaging barriers [1,2]. Environmental concerns and regulatory shifts have accelerated compostable material development to replace conventional petroleum-based plastics [3,4]. Because many active packaging technologies rely on persistent synthetic polymers, sustainable materials must deliver robust food protection and end-of-life environmental compatibility.

Polyvinyl alcohol (PVOH) is a water-soluble polymer recognized for its film-forming capability, strong substrate adhesion, and superior oxygen barrier performance under dry conditions [5,6]. The high density of backbone hydrox-

yl groups promotes extensive hydrogen bonding, creating a tightly packed crystalline network that restricts gas diffusion [5]. The PVOH supports water-based processing and biodegrades under appropriate environmental conditions, making it an excellent sustainable packaging coating matrix [5,6]. However, neat PVOH lacks intrinsic antimicrobial activity and requires active agents.

Among metal-based active agents, copper is suited for food packaging due to its cost-effectiveness, natural abundance, and broad-spectrum antimicrobial utility [7,8]. Copper ions deactivate pathogens through multiple mechanisms — including cell membrane disruption, reactive oxygen species generation, and enzyme interference — which significantly reduces the likelihood of microbial resistance compared to conventional organics [9,10]. While effective, copper nanoparticles raise concerns regarding

migration, environmental persistence, and regulatory compliance [7,9]. Ionic copper systems offer a simpler, commercially favorable approach for food contact materials [7,8].

Embedding copper ions within PVOH combines antimicrobial defenses with sustainable coating design. Because active additives must not hinder end-of-life processing, evaluating industrial compostability is important [11,12]. However, limited research has systematically investigated copper ion-embedded PVOH coatings across the interconnected dimensions of biological efficacy, industrial biodegradation, and physical barrier performance.

This study aimed to develop and characterize an active, compostable copper ion-embedded PVOH coating for food-packaging applications. The PVOH formulations containing 100, 500, and 1,000 ppm Cu^{2+} were applied to paper and polylactic acid (PLA) substrates at a target weight of 6 g/m². The resulting films were evaluated for antimicrobial efficacy against *Escherichia coli* and *Salmonella* spp.; industrial compostability; and comprehensive thermal, barrier, morphological, rheological, and surface properties.

MATERIALS AND METHODS

Materials

Polyvinyl alcohol powder with an 88% degree of hydrolysis and ~95% polymer solids (Kuraray America; Houston, TX, USA) served as the base polymer. Anhydrous copper(II) chloride (CuCl_2 , Sigma-Aldrich; St. Louis, MO, USA) served as the copper ion source. Food-grade lactic acid (Sigma-Aldrich) was used for pH adjustment, and a commercial defoaming agent (FC 407, ESP Enterprises; Houston, TX, USA) was incorporated during solution preparation. Deionized water was used throughout.

Commercial PLA film (Evlon grade, BI-AX International; Wingham, ON, Canada) with a nominal thickness of 40 μm served as the polymeric substrate. Unbleached, uncoated butcher paper and parchment paper served as representative paper-based food packaging substrates.

For antimicrobial assays, *Escherichia coli* O157 and *Salmonella* spp. served as representative foodborne pathogens. Standard microbiological media, including nutrient agar and soft agar overlays (0.8% agar), were prepared according to standard laboratory protocols.

Design of copper-PVOH coating formulations

Copper-PVOH coating formulations maintained constant polymer solids content while systematically varying copper ion concentration. All formulations were prepared on a mass basis to improve reproducibility. Target PVOH solids content was fixed at 20% (w/w), accounting for volatile content in the polymer powder.

Copper ion concentrations of 0 (control), 100, 500, and 1,000 ppm Cu^{2+} (mass basis) were selected to evaluate concentration-dependent effects on coating performance. A copper-free 20% PVOH formulation served as the reference control for all comparative analyses.

Preparation of copper-PVOH coating solutions

The PVOH coating solutions were prepared by gradually adding PVOH powder to deionized water under continuous mechanical mixing using a rotor-stator mixer (POLYTRON PT 2100, Kinematica AG; Malers, Switzerland). A defoaming agent was incorporated at 1 wt% of the formulation during mixing. The mixture was heated to 90°C–95°C under continuous mixing until complete dissolution, yielding a clear, homogeneous solution. The solution was then cooled to ambient temperature under continuous stirring.

Anhydrous CuCl_2 was incorporated by direct gravimetric addition or dilution, depending on target copper concentration, to ensure accurate and reproducible copper loading. Copper-containing solutions were mixed thoroughly for uniform copper ion distribution and equilibrated before coating application. Formulation pH was maintained between 4.5 and 5.5 using food-grade lactic acid. This pH range was selected to promote dissolved Cu^{2+} ion stability and minimize copper hydroxide formation, thereby maintaining copper availability within the coating solution prior to application.

Coating application and sample conditioning

Copper-PVOH coatings were applied to PLA films, butcher paper, and parchment paper using a laboratory coating unit with a No. 11 coating roller. Coating parameters were controlled to achieve a target dry coating weight of 6 g/m².

After application, coated samples were dried under ambient laboratory conditions of $23 \pm 2^\circ\text{C}$ and $50 \pm 5\%$ relative humidity (RH) for 24 h and conditioned for an additional 24–36 h before testing. Uncoated substrates and copper-free PVOH-coated substrates were prepared and conditioned identically for comparison.

Biodegradability evaluation under industrial composting conditions

Biodegradability of copper-embedded PVOH-coated butcher paper and uncoated butcher paper was evaluated under controlled industrial composting conditions in accordance with ASTM D5338 “Standard test method for determining aerobic biodegradation of plastic materials under controlled composting conditions, incorporating thermophilic temperatures” and ASTM D6400 “Standard specification for labeling of plastics designed to be aerobically composted in municipal or industrial facilities.”

Composting experiments used a laboratory-scale composting setup maintained at $58 \pm 2^\circ\text{C}$ under aerobic conditions. Test materials were cut into small pieces, dried to determine initial dry mass, and mixed with mature compost. Blank compost reactors accounted for background carbon dioxide (CO_2) evolution.

Carbon dioxide generated during composting was periodically collected and quantified. Cumulative CO_2 evolution from test samples was corrected using compost blanks to evaluate biodegradation behavior over the test duration.

Provisions remain for incorporating additional biodegradation data and tables upon completion of testing.

Antimicrobial performance evaluation

Antimicrobial activity of copper-embedded PVOH coatings was evaluated using copper ion concentrations of 0 ppm (control), 100 ppm, and 1,000 ppm Cu^{2+} . Testing was performed directly on the coating solution, coated parchment paper, and coated PLA film (6 g/m² dry coating). Copper-free coatings and uncoated substrates served as negative controls.

A bacterial overlay agar method assessed antimicrobial efficacy. Test specimens were placed on a solidified base agar layer, followed by application of a soft agar overlay inoculated with *Escherichia coli* or *Salmonella* spp. The plates were incubated at 37°C for 24 h under aerobic conditions. Antimicrobial performance was qualitatively evaluated by visually observing bacterial growth suppression directly above or around the specimens as compared with controls.

Thermal characterization by differential scanning calorimetry

Thermal behavior of copper-embedded PVOH coatings was characterized by differential scanning calorimetry (DSC) using the DSC 2500 from TA Instruments (New Castle, DE, USA). Neat PVOH and copper-containing PVOH samples with 100, 500, and 1,000 ppm Cu^{2+} were analyzed for concentration-dependent thermal behavior.

Samples underwent a heat-cool-reheat program at a constant heating and cooling rate of 10°C/min under nitrogen. Glass transition temperature (T_g) and melting temperature (T_m) were determined from the second heating cycle, while crystallization temperature (T_c) was obtained from the cooling segment.

Permeability performance evaluation

Commercial PLA films (40- μm nominal thickness) served as substrates for permeability testing. The 1,000-ppm Cu^{2+} -embedded PVOH formulation was applied to the PLA films via laboratory drawdown at a target dry coating weight of 6 g/m². Coated samples were dried under ambient laboratory conditions (23 $\pm$ 2°C, 50 $\pm$ 5% RH) for 24 h and conditioned for an additional 24–36 h before characterization. Uncoated PLA films served as controls.

Barrier performance was assessed through oxygen transmission rate (OTR) and water vapor transmission rate (WVTR) measurements. The OTR was measured using a MOCON OX-TRAN system (AMETEK; Berwyn, PA, USA) in accordance with ASTM D3985 “Standard test method for oxygen gas transmission rate through plastic film and sheeting using a coulometric sensor,” with steady-state values reported in cubic centimeters per one hundred square inches per day (cc/100 in²-day). The WVTR was measured using a MOCON PERMATRAN system (AMETEK) in accor-

dance with ASTM F1249 “Standard test method for water vapor transmission rate through plastic film and sheeting using a modulated infrared sensor,” with equilibrium values reported in grams per one hundred square inches per day (g/100 in²-day).

Contact angle analysis

Water contact angle measurements evaluated surface wettability of uncoated and Cu-PVOH-coated PLA films. The Cu-PVOH formulation was applied to the PLA films (40- μm thickness) using a laboratory coating unit with a number 12 metering rod for a target dry coating weight of 6 g/m². Following ambient drying, both coated and uncoated substrates were sectioned into 3 in. $\times$ 3 in. specimens for characterization.

Surface wettability was quantified using a DSA 10MK2 drop shape analysis system (Krüss Scientific; Hamburg, Germany) via the sessile drop method with deionized water as probe liquid. A controlled droplet was deposited on the film surface, and the contact angle was tracked over 60 s to observe wetting dynamics. Replicate measurements at multiple distinct locations on each specimen determined average values.

Viscosity measurement of Cu-PVOH coating formulations

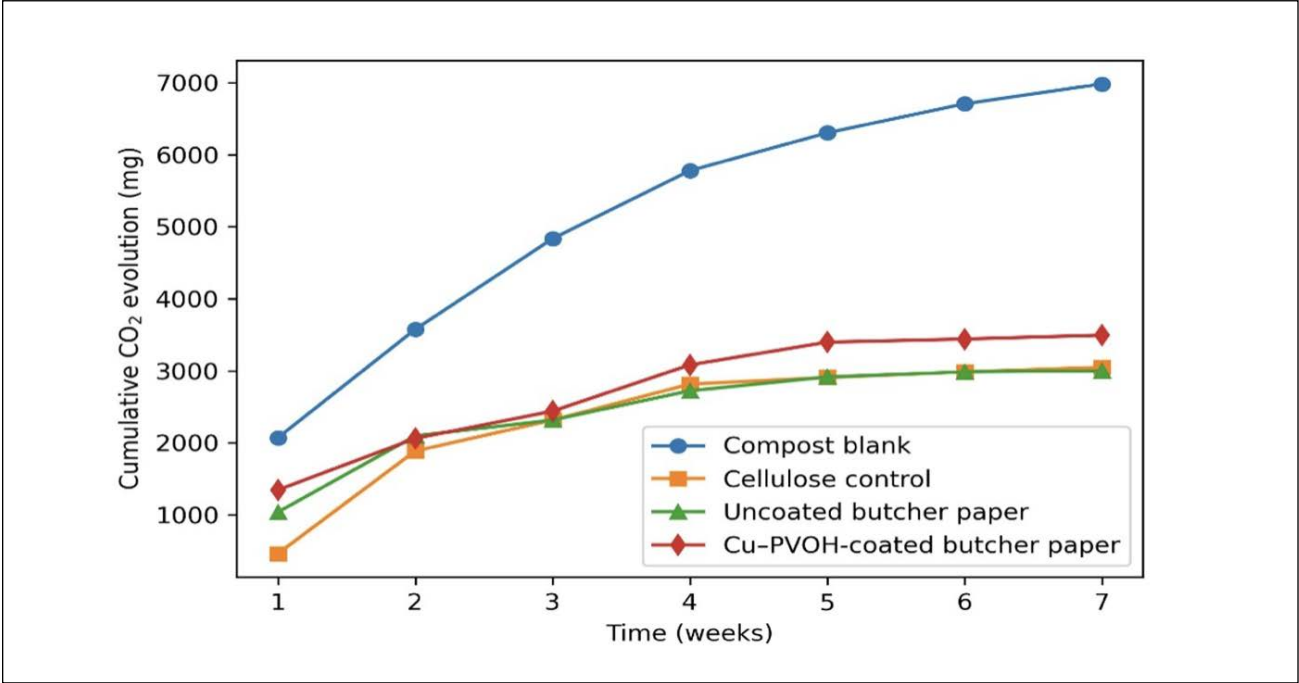
Rheological behavior of Cu-PVOH coating formulations was evaluated using a Brookfield digital rotational viscometer (Model LVDV-E, Brookfield Engineering Laboratories; Middleborough, MA, USA). Viscosity measurements assessed the influence of copper ion concentration on coating-solution flow behavior at a constant PVOH solids content of 20% (w/w).

Four formulations were evaluated: a copper-free PVOH control and Cu-PVOH formulations containing 100, 500, and 1,000 ppm Cu^{2+} . Before characterization, each formulation was gently stirred to ensure homogeneity while minimizing air bubble incorporation. Measurements used an LV-2 (62) spindle at 30 rpm and approximately 25°C. Viscosity was recorded after stabilization and reported in millipascal-seconds (mPa·s).

Morphological analysis

Surface morphology and elemental composition of Cu-PVOH coatings were analyzed using a Hitachi S-3400N scanning electron microscope (SEM) equipped with an energy dispersive x-ray spectroscopy (EDS) detector (Hitachi High-Tech; Tokyo).

Samples included a control PVOH film (0 ppm Cu^{2+}); solvent-cast PVOH films containing 500 ppm and 1,000 ppm copper ions; and a 1,000 ppm Cu-PVOH coating applied to a 40 μm PLA substrate at a dry coating weight of 6 g/m². Free-standing films were prepared via solvent casting under ambient laboratory conditions. The coated PLA sample was produced under controlled coating conditions to



1. Weekly cumulative carbon dioxide (CO₂) evolution during ASTM D5338 industrial composting of uncoated butcher paper and copper ion-embedded polyvinyl alcohol (Cu-PVOH)-coated butcher paper (1000 ppm Cu, 6 g/m²) compared with compost blank and cellulose control at 58 ± 2°C.

achieve uniform coverage. Samples were mounted on aluminum stubs using conductive carbon tape and examined under high-vacuum conditions. The EDS analysis confirmed copper incorporation and relative elemental content.

RESULTS AND DISCUSSION

Biodegradability performance under industrial composting conditions

The biodegradability of copper-embedded PVOH-coated substrates was evaluated under controlled industrial composting conditions in accordance with ASTM D5338. **Figure 1** presents the cumulative CO₂ evolution profiles for uncoated butcher paper and paper coated with 1,000 ppm Cu-PVOH at a coating weight of 6 g/m² over a 49-day composting period. The CO₂ evolution curves were corrected for background compost respiration using blank controls.

Uncoated butcher paper exhibited a gradual increase in CO₂ evolution throughout the composting period, reaching 71.90% biodegradation by the end of the test. In contrast, Cu-PVOH-coated paper achieved 83.59% biodegradation under identical conditions, indicating that the coating did not inhibit microbial activity during composting.

The enhanced biodegradation observed for the coated samples may be attributed to the hydrophilic nature of PVOH, which facilitates moisture uptake and microbial accessibility to both the coating and the underlying paper substrate [5,11]. Importantly, incorporation of copper ions at the tested concentration did not adversely affect biodegradation, suggesting that the copper loading was sufficient-

ly low to avoid inhibition of compost microorganisms while remaining effective against foodborne pathogens during antimicrobial testing [13]. **Table I** summarizes the final biodegradation values for the tested materials.

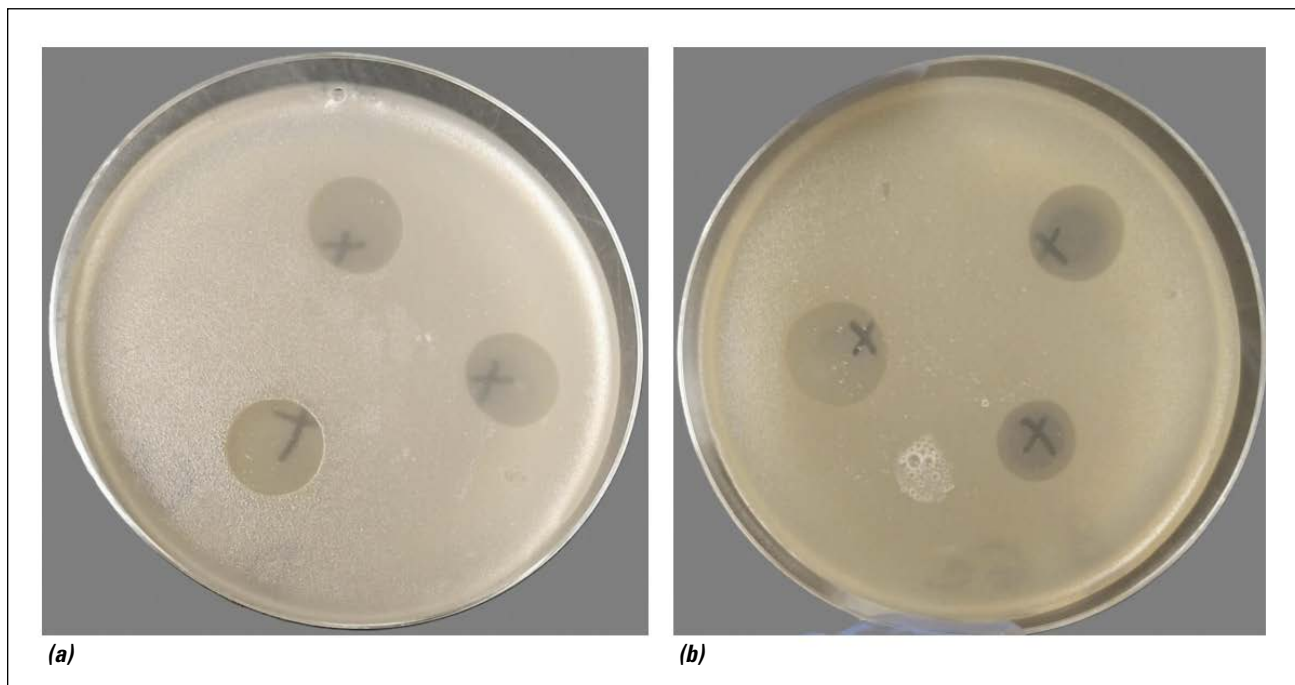
The results demonstrate that copper-embedded PVOH coatings are compatible with industrial composting systems and may enhance biodegradation relative to uncoated paper. Maintaining biodegradation performance while introducing antimicrobial functionality addresses a critical challenge in active packaging design and supports the potential use of Cu-PVOH coatings in sustainable food-packaging applications.

Antimicrobial performance of Cu-PVOH coatings

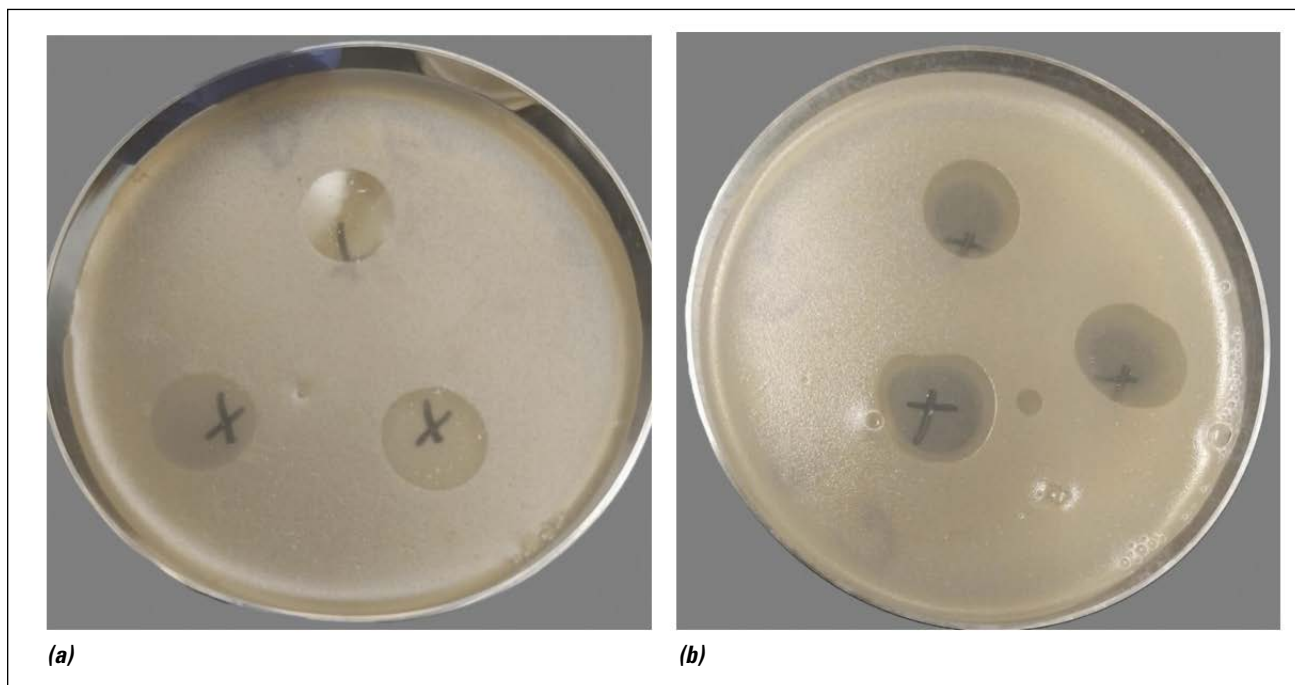
The antimicrobial response of Cu-PVOH systems demonstrated a clear dependence on copper concentration and substrate characteristics. The results indicate that copper

Sample	Biodegradation, %
Cellulose (reference)	72.88
Uncoated butcher paper	71.90
PVOH + Cu coated butcher paper	83.59

1. Final percent biodegradation of uncoated butcher paper and copper ion-embedded polyvinyl alcohol (Cu-PVOH)-coated paper (1,000 ppm Cu, 6 g/m²) after 49 days of industrial composting (ASTM D5338, 58 ± 2°C).



2. Antimicrobial activity of Cu-PVOH coatings containing (a) 100 ppm Cu^{2+} and (b) 1,000 ppm Cu^{2+} against *Escherichia coli*.

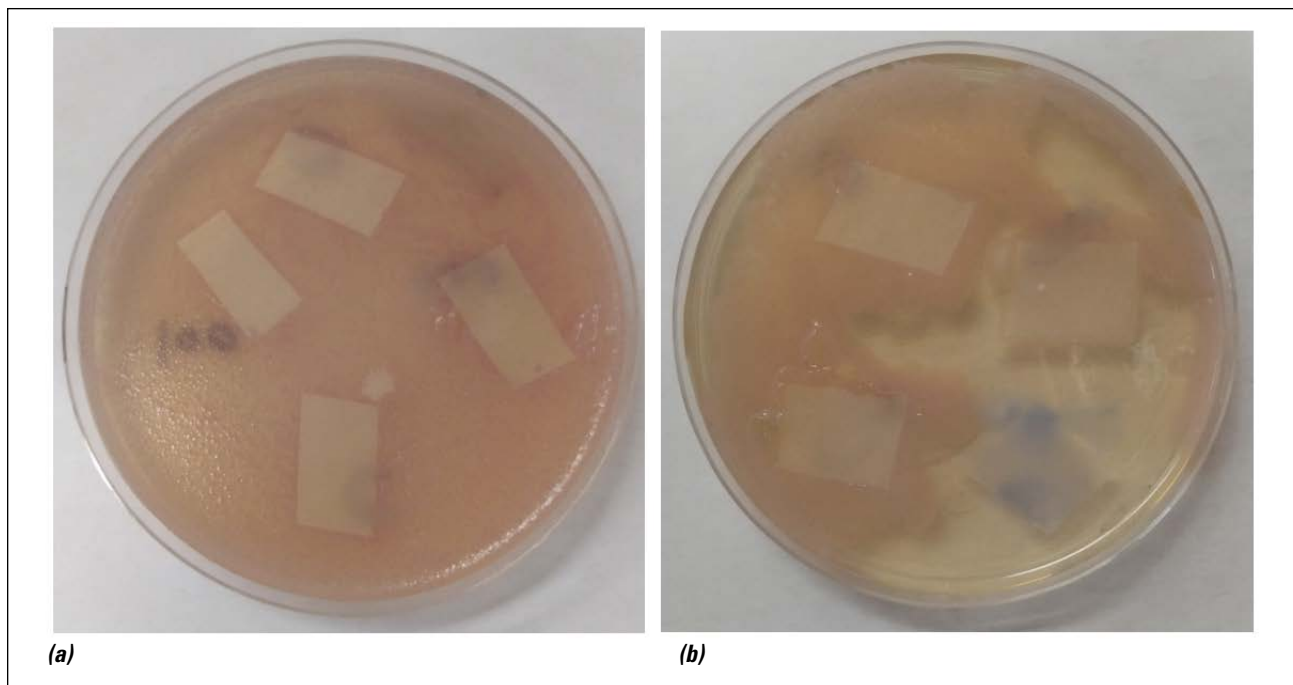


3. Antimicrobial activity of Cu-PVOH coatings containing (a) 100 ppm Cu^{2+} and (b) 1,000 ppm Cu^{2+} against *Salmonella* spp.

ion availability and diffusion behavior play important roles in determining antimicrobial effectiveness [7].

When evaluated in solution form, both 100 ppm and 1,000 ppm Cu^{2+} concentrations exhibited clear inhibition of *Escherichia coli* and *Salmonella* spp., as evidenced by visible growth suppression corresponding to the coating application areas. As shown in **Fig. 2** and **Fig. 3**, micro-

bial inhibition was primarily confined to the regions where the Cu-PVOH coating was directly present, with no significant inhibition extending beyond these boundaries. Similar inhibition behavior was observed for both bacterial species, indicating that the antimicrobial activity of the Cu-PVOH system was effective against both gram-negative foodborne pathogens evaluated in this study.



4. Antimicrobial activity of Cu–PVOH-coated parchment paper containing (a) 100 ppm Cu²⁺ and (b) 1,000 ppm Cu²⁺ against *Escherichia coli*.

The localized nature of the inhibition suggests that copper ions remained predominantly associated with the PVOH matrix rather than diffusing extensively into the surrounding agar medium. This behavior may be advantageous for food-packaging applications because antimicrobial activity is concentrated at the package surface, where microbial contamination is most likely to occur, while potentially reducing unnecessary migration of active components into the surrounding environment [7,9].

In contrast, substrate-coated samples showed reduced antimicrobial performance, particularly at lower copper loading. As shown in **Fig. 4**, the 100 ppm Cu²⁺ coating on parchment paper exhibited no visible inhibition, whereas the 1,000 ppm Cu²⁺ coating demonstrated only partial inhibition against *Escherichia coli*. This reduced effectiveness can be attributed to the porous and fibrous structure of parchment paper, which likely promoted copper ion penetration into the substrate and reduced the surface concentration available for antimicrobial action.

The antimicrobial behavior of Cu–PVOH coatings on PLA film differed from that of parchment paper because of the non-porous nature of the PLA substrate. The PLA samples coated with 100 ppm and 1,000 ppm Cu²⁺ showed clear bacterial inhibition, localized to the coating application regions, against *Escherichia coli*, as shown in **Fig. 5**. The absence of extended inhibition zones suggests that copper ions remained primarily associated with the coating surface rather than diffusing into the surrounding agar medium [7].

The presence of inhibition at both copper concentrations indicates that, on a non-absorptive substrate such as PLA, even lower copper loadings can provide sufficient

surface-available copper for antimicrobial performance. This localized response is advantageous for food-packaging applications because it supports contact-active microbial inhibition while minimizing unnecessary ion migration [8].

Overall, antimicrobial performance followed the trend: solution > PLA substrate > parchment paper. While both 100 ppm and 1,000 ppm Cu²⁺ exhibited effective inhibition in solution, antimicrobial activity in coated systems depended on substrate morphology and the surface availability of copper ions. Inhibition was primarily localized to the coating application regions, indicating a contact-active mechanism with minimal ion diffusion. Further studies need to be performed to establish if Cu²⁺ migrates from the film and, if so, in what amount.

Thermal behavior of copper ion-embedded PVOH coatings

Differential scanning calorimetry was used to evaluate the influence of copper-ion incorporation on the thermal behavior of PVOH coatings. Heating and cooling cycles were analyzed to assess changes in thermal transitions with increasing copper concentration.

DSC heating behavior

The DSC heating curves of neat PVOH and copper ion-embedded PVOH coatings are presented in **Fig. 6**. All samples exhibited characteristic PVOH thermal features, including a distinct glass transition region followed by a melting endotherm at elevated temperatures [14].

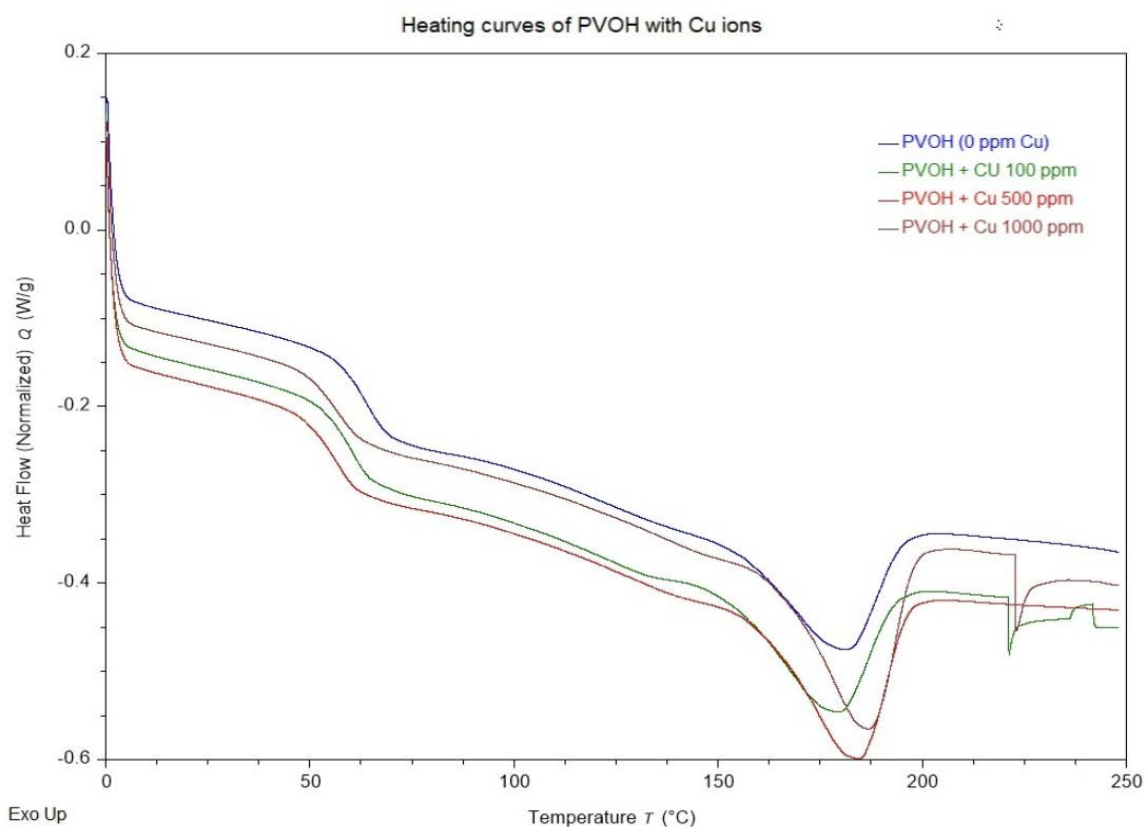
Compared with neat PVOH, the copper-containing formulations showed distinct shifts in thermal transitions.



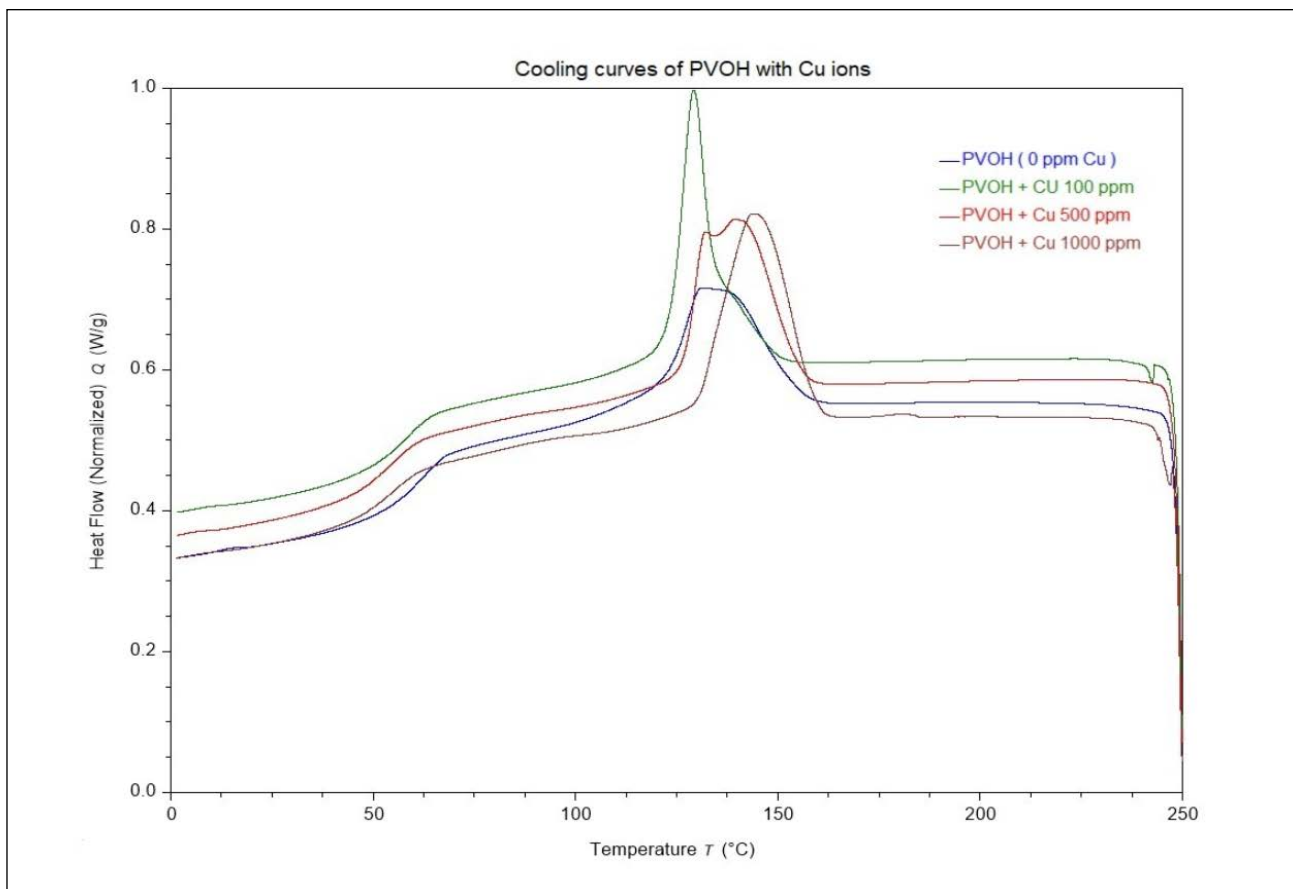
(a)

(b)

5. Antimicrobial response of Cu-PVOH-coated PLA film containing (a) 100 ppm Cu^{2+} and (b) 1,000 ppm Cu^{2+} against *Escherichia coli*.



6. Differential scanning calorimetry (DSC) heating curves of neat PVOH and Cu-PVOH coatings containing 100, 500, and 1,000 ppm Cu, illustrating changes in thermal transitions as a function of copper ion concentration.



7. The DSC cooling curves of neat PVOH and copper-ion-embedded PVOH coatings containing 100, 500, and 1,000 ppm Cu, showing crystallization behavior during controlled cooling.

These variations are attributed to the formation of coordination complexes between Cu^{2+} ions and backbone hydroxyl groups, which restrict polymer chain mobility and alter crystalline domain organization [15,16]. At higher copper loadings, the melting endotherm broadened, reflecting changes in crystal size distribution. However, all coatings retained well-defined melting profiles, confirming that copper incorporation up to 1,000 ppm did not compromise the fundamental thermal stability of the PVOH system.

DSC cooling behavior and crystallization characteristics

The DSC cooling curves of neat PVOH and copper ion-embedded PVOH coatings are shown in **Fig. 7**. During cooling, all samples exhibited exothermic crystallization peaks associated with polymer chain reorganization and crystal formation [15,16]. Copper incorporation influenced both the position and intensity of the crystallization exotherm, indicating changes in crystallization behavior.

At lower copper concentrations, the crystallization response reflects heterogeneous nucleation effects, while higher copper loading broadens the crystallization peak, confirming restricted chain mobility during cooling [15]. These observations are consistent with copper-polymer coordination complexes that alter the kinetics and extent

of crystalline phase development. Overall, the heating and cooling results indicate that copper ion incorporation modifies the thermal response of PVOH without compromising its thermal integrity. This supports the suitability of Cu-PVOH coatings for coating formation, drying, and food-packaging applications.

Permeability performance of Cu-PVOH-coated PLA films

The OTR and the WVTR of uncoated and Cu-PVOH-coated (1,000 ppm Cu^{2+}) PLA films were evaluated at a dry coating weight of 6 g/m² in accordance with ASTM D3985 and ASTM F1249, respectively. Table II summarizes the permeability test comparison of Cu-PVOH coating with PLA film.

The Cu-PVOH coating substantially improved oxygen barrier performance, with OTR decreasing sharply from 23.01 to 0.10 cc/100 in²-day. This reduction is attributed to the dense, hydrogen-bonded structure of the PVOH matrix, which restricts oxygen diffusion [6]. The copper incorporation did not disrupt the coating's structural continuity. Because the formulation utilized an 88% degree of hydrolysis (DoH) PVOH, even lower oxygen permeability may be achievable with higher-DoH grades due to increased crystallinity and stronger intermolecular bonding [15,16].

Sample	OTR, cc/100 in ² -day	WVTR, g/100 in ² -day
Uncoated PLA	23.01	11.98
Cu-PVOH-coated PLA	0.10	11.91

II. Oxygen transmission rate (OTR) and water vapor transmission rate (WVTR) of uncoated and Cu-PVOH-coated polylactic acid (PLA) films.

In contrast, WVTR values were essentially unchanged, shifting only from 11.98 to 11.91 g/100 in²-day. This negligible difference confirms that the coating did not enhance water-vapor barrier properties, which is consistent with the hydrophilic nature of PVOH and its strong interaction with moisture [5,6]. Overall, these results demonstrate the suitability of Cu-PVOH coatings for packaging applications where oxygen barrier performance is the primary requirement.

Contact angle analysis

Water contact angle measurements were performed to evaluate the surface wettability of uncoated PLA and Cu-PVOH-coated PLA films. The uncoated PLA exhibited an average contact angle of 69.0°, while the Cu-PVOH-coated PLA formulation exhibited an average contact angle of 71.0°. This minimal change indicates that the application of the Cu-PVOH layer did not substantially alter the macrostructural surface wettability of the underlying PLA substrate.

Although neat PVOH is inherently hydrophilic and typically exhibits a much lower contact angle, the cross-linking effect or coordination complexes formed between the incorporated copper ions and the PVOH hydroxyl groups effectively reduced free hydrophilic sites at the interface [5,6]. This structural interaction, combined with the low coating weight, maintained a balanced surface polarity. These results confirm that copper incorporation does not adversely affect the surface characteristics of the composite film, maintaining a surface wettability profile that is highly suitable for subsequent industrial printing, sealing, and converting operations.

Viscosity behavior of Cu-PVOH coating formulations

The viscosity of the control PVOH and copper-embedded PVOH formulations (100, 500, and 1,000 ppm Cu²⁺) was measured using a Brookfield LV-DVE viscometer (spindle 62, 30 rpm) at approximately 25°C. All formulations were prepared at 20% solids content. **Table III** presents measured viscosity values.

Viscosity values ranged from 917 to 942 mPa·s, showing no significant variation among formulations. The incorporation of copper ions up to 1,000 ppm did not substantially affect the rheological behavior of the 20% PVOH system.

All formulations exhibited viscosities within a suitable

Formulation	Viscosity, mPa·s
PVOH Control	923
PVOH + 100 ppm Cu	917
PVOH + 500 ppm Cu	942
PVOH + 1000 ppm Cu	931

III. Viscosity of Cu-PVOH coating formulations.

range for coating application, indicating that copper loading at the studied levels does not compromise coating processability or solution stability.

Morphological analysis of Cu-PVOH coatings

Surface morphology with SEM

Micrographs from scanning electron microscopy (SEM) are presented in **Fig. 8**. The control PVOH film exhibited a smooth and homogeneous surface without visible particulate features, indicating uniform film formation.

Incorporation of copper ions resulted in noticeable morphological changes. The 500-ppm Cu-PVOH film displayed dispersed microscale domains within the polymer matrix, attributed to copper distribution. Increasing the copper concentration to 1,000 ppm resulted in a higher density of these features and greater surface heterogeneity, similar to observations reported for copper-containing PVA composites [17].

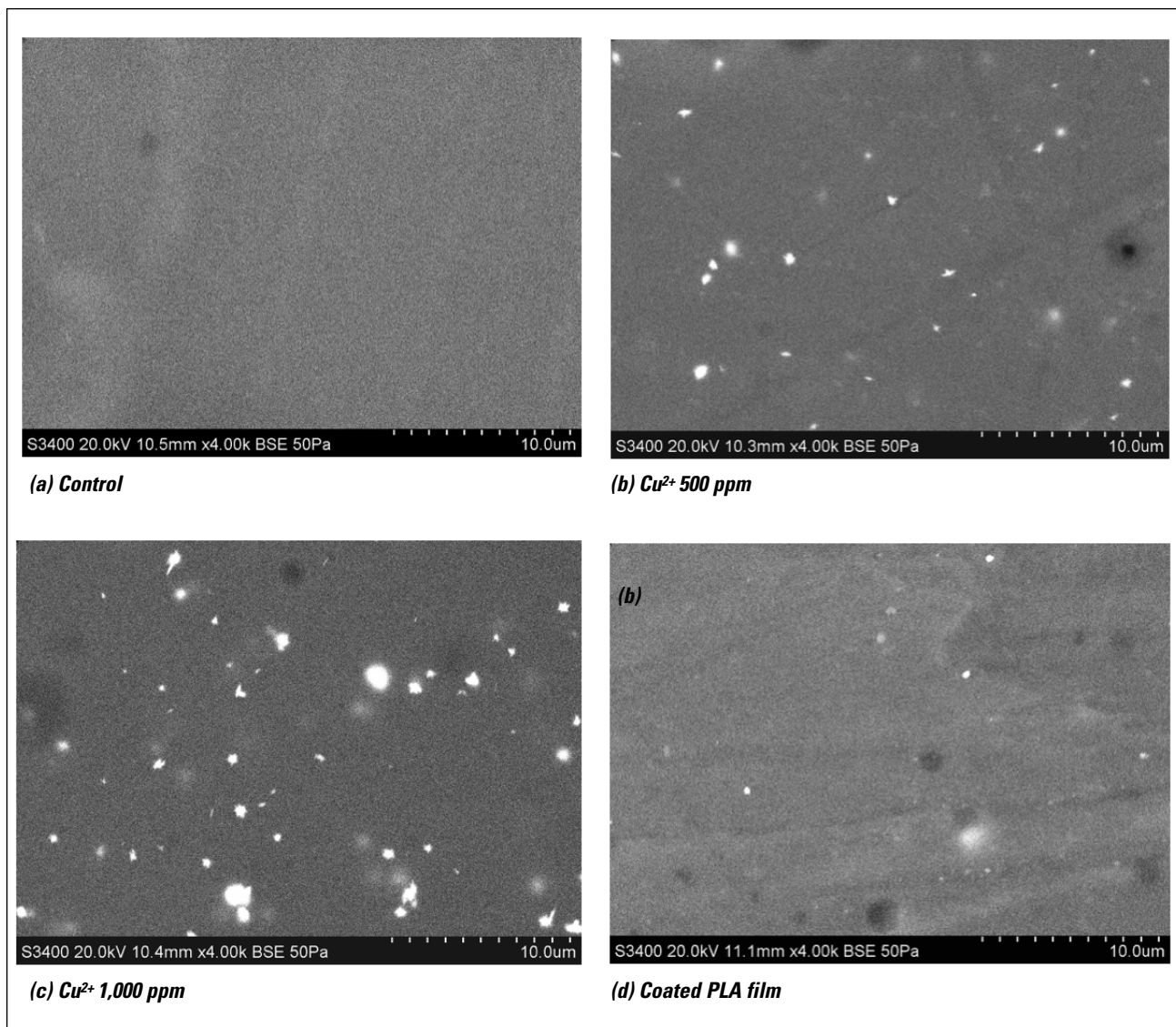
The 1,000-ppm Cu-PVOH-coated PLA sample exhibited a continuous coating layer over the substrate surface. Embedded features consistent with copper incorporation were observed while maintaining uniform coverage of the PLA film.

Overall, surface heterogeneity increased with copper concentration, confirming successful integration of copper within the PVOH matrix. The absence of major surface defects suggests good compatibility between the PVOH matrix and incorporated components [18].

Elemental analysis with EDS

Spectra from EDS are presented in **Fig. 9**. No copper peak was detected in the control PVOH film, confirming the absence of copper in the baseline formulation. The 500-ppm Cu-PVOH film exhibited a measurable copper peak corresponding to approximately 0.3 wt% copper. Increasing the copper concentration to 1,000 ppm resulted in a stronger and more pronounced copper peak, corresponding to approximately 1.0 wt%.

The 1,000-ppm Cu-PVOH-coated PLA sample exhibited a copper signal of approximately 0.8 wt%, slightly lower than that of the free-standing 1,000 ppm film due to contributions from the underlying PLA substrate and the limited coating thickness (6 g/m²). The progressive increase in



8. Micrographs from scanning electron microscopy (SEM) of Cu-PVOH coatings and coated PLA substrate: (a) control; (b) 500 ppm Cu^{2+} ; (c) 1,000 ppm Cu^{2+} ; and (d) PLA film coated with Cu-PVOH containing 1,000 ppm Cu^{2+} .

copper peak intensity with increasing formulation concentration validates successful copper incorporation within the coating system. Similar elemental confirmation of copper incorporation in PVA-based composites has been reported using EDS characterization techniques [19].

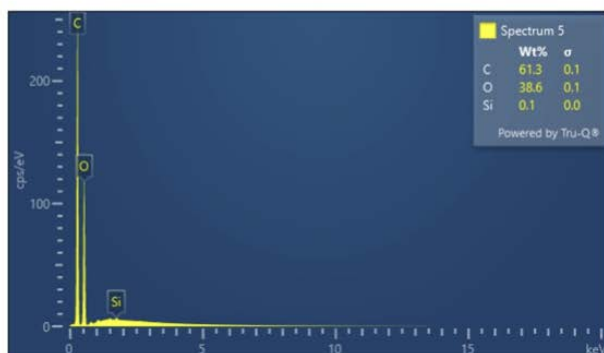
During SEM analysis, rapid matrix degradation occurred under prolonged electron beam exposure due to chain scission. However, this localized organic mass loss did not compromise the relative elemental quantification of the embedded copper ions.

CONCLUSIONS

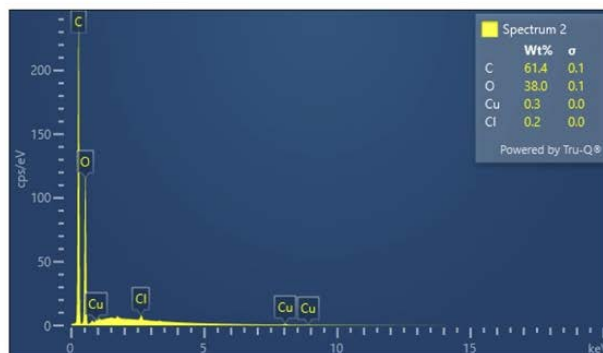
This study successfully developed and characterized active, compostable copper ion-embedded PVOH coatings tailored for sustainable food-packaging applications. The antimicrobial efficacy of the formulations was strongly dependent on

the copper ion concentration and substrate morphology, following the performance trend of solution > PLA film > parchment paper. Crucially, the antimicrobial inhibition was predominantly confined to the direct surface contact zones with minimal ion diffusion. This contact active mechanism provides targeted food protection while minimizing unnecessary metallic migration into the food matrix or surrounding environment.

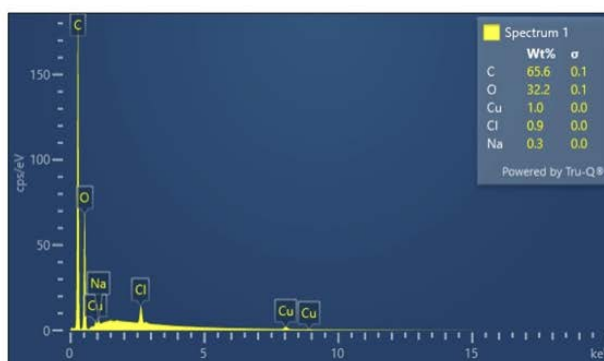
Biodegradability assessments verified that the incorporation of up to 1,000 ppm copper ions did not hinder or delay macromolecular decomposition. The copper-embedded PVOH coatings remained completely compatible with industrial composting loops, with the coated paper achieving a high 83.59% biodegradation over a 49-day cycle under ASTM D5338 and ASTM D6400 protocols. Furthermore, the functional performance of the underlying substrates was



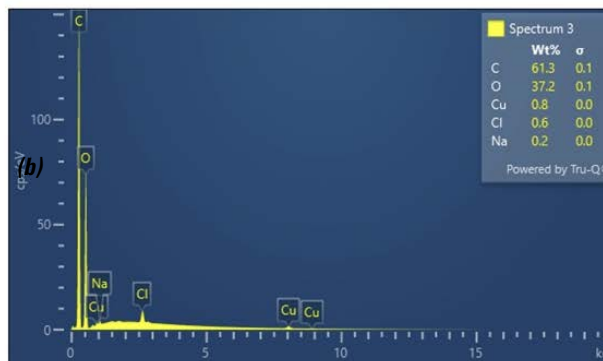
(a) Control



(b) Cu-PVOH 500 ppm



(c) Cu-PVOH 1,000 ppm



(d) Cu-PVOH coated PLA film

9. Spectra from energy dispersive x-ray spectroscopy (EDS) of Cu-PVOH coatings and coated PLA substrate: (a) control; (b) 500 ppm Cu²⁺; (c) 1,000 ppm Cu²⁺; and (d) PLA film coated with Cu-PVOH containing 1000 ppm Cu²⁺.

substantially improved; the application of the continuous coating layer achieved an exceptional reduction in oxygen transmission rate from 23.01 to 0.10 cc/100 in²-day. Conversely, water vapor transmission rates remained statistically unchanged due to the PVOH matrix's inherent hydrophilicity.

Finally, rheological, thermal, and surface contact angle evaluations demonstrated that copper loading did not compromise the industrial processability, solution stability, or macrostructural film integrity of the 20% solids content polymer system. In conclusion, copper ion-embedded PVOH coating technology offers paper, bioplastic, and bio-based packaging converters a viable, high-performance manufacturing strategy to integrate robust antimicrobial defenses into functional, commercially compostable packaging formats.

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This research aimed to develop an antimicrobial and biodegradable coating with the potential to reduce food loss and plastic waste, supporting more sustainable food packaging. Our study extends previous antimicrobial packaging research by combining antimicrobial performance, biodegradability, and potential industrial coating application in a PVOH–copper system.

The main challenge in this study was balancing antimicrobial efficacy with biodegradability. This was addressed by evaluating copper ion concentrations within the PVOH coating.

The PVOH–copper coating demonstrated very good inhibition of common food-borne pathogens while retaining biodegradability under industrial composting conditions. Some interesting findings were that the PVOH–copper coating did not hinder biodegradation of butcher paper and that coated samples showed greater biodegradation than uncoated samples under the tested composting conditions.

Mills and converters could use this concept to develop antimicrobial, biodegradable food-packaging coatings using PVOH and copper salts with potential compatibility with industrial coating processes.



Ranil



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Future work should quantify copper migration and evaluate antimicrobial efficacy in real food systems, considering moisture, pH, and packaging atmosphere.

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