



## RESEARCH ARTICLE

PREPARATION AND CHARACTERIZATION OF PVA/CELLULOSE NANOCRYSTALS/ $\epsilon$ -POLY-L-LYSINE BIOCOMPOSITES FOR POST-HARVEST PRESERVATION OF CHILLIES

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**Abstract.** Post-harvest deterioration of highly perishable product, such as chillies, remains a major concern in global food supply chains, resulting in huge economic losses and food waste, as well as highlighting the need for sustainable alternatives to synthetic preservatives. This work created multifunctional biocomposite coatings using polyvinyl alcohol (PVA), cellulose nanocrystals (CNC), and  $\epsilon$ -polylysine ( $\epsilon$ -PL) to improve mechanical, barrier, and antibacterial properties. The films were created using high-shear homogenisation and thoroughly characterised with optical microscopy (OM), Fourier transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM), and X-ray diffraction (XRD). OM and SEM investigations showed uniform dispersion at ideal compositions and aggregation at greater  $\epsilon$ -PL loadings. FTIR demonstrated significant intermolecular interactions via hydrogen bonding and electrostatic effects. XRD results show a composition-dependent balance between CNC-induced crystallinity and  $\epsilon$ -PL-induced disruption of ordered domains, highlighting the importance of additive concentration in determining film structure. The biocomposite coatings were applied to fresh chillies and tested for 21 days under ambient storage conditions. The optimised formulation with 3 wt.%  $\epsilon$ -PL reduced weight loss to 1.70 %, compared to 9.52 % for the control and 5.11 % for neat PVA. It also maintained visual quality and stiffness. The improved performance is attributable to the creation of a compact, well-integrated network structure that improves moisture barrier qualities while also providing bioactive functionality. This study establishes a clear structure property performance relationship and demonstrates the potential of PVA/CNC/ $\epsilon$ -PL biocomposites as scalable, environmentally friendly coatings for post-harvest preservation and active food packaging applications.

**Keywords:** Biocomposites, chillies, post-harvest preservation, cellulose nanocrystals (CNC), polyvinyl alcohol (PVA).

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## 1. INTRODUCTION

Fresh produce post-harvest deterioration is still a significant problem in global food supply chains, especially for highly perishable goods like chillies, which are prone to moisture loss, microbial spoiling, and quick quality degradation during storage and transit. Conventional packaging materials, which are mostly made of synthetic polymers that are not biodegradable, are problematic for the environment and frequently do not have the combination barrier and antibacterial properties needed for efficient preservation. As a result, there is growing interest in creating multipurpose, biodegradable, and sustainable packaging technologies that can preserve product quality while lessening their negative effects on the environment [1].

Because biopolymer-based edible coatings may provide thin, protective layers that control microbial growth, gas exchange, and moisture transfer, they have become attractive substitutes. Due to its superior film-forming capacity, biocompatibility, and low toxicity, polyvinyl alcohol (PVA) is widely employed; yet, its intrinsic barrier and mechanical qualities are still restricted. Because of their high crystallinity, vast surface area, and capacity to function as efficient reinforcing nanofillers, cellulose nanocrystals (CNC), which are generated from lignocellulosic biomass, have been demonstrated to improve these qualities [2]. Furthermore, CNC can reduce gas and moisture permeability by forming a convoluted diffusion pathway within the polymer matrix [3]. In the meanwhile,  $\epsilon$ -poly-L-lysine ( $\epsilon$ -PL), a naturally occurring antimicrobial peptide that has been licensed for use in food applications, offers active activity by suppressing spoilage germs by electrostatically breaking microbial cell membranes [4].

Despite these developments, research has mostly concentrated on either antibacterial functionality or reinforcement, with little knowledge of their combined integration inside a single matrix. Specifically, not enough research has been done on the synergistic interactions between CNC and  $\epsilon$ -PL and how they affect the structure-property-performance relationship of biocomposite coatings. Furthermore, the application of many published techniques in actual post-harvest preservation is limited due to their lack of thorough evaluation under realistic storage circumstances.

To address these gaps, this study introduces a multifunctional PVA/CNC/ $\epsilon$ -PL biocomposite coating that simultaneously integrates nanoreinforcement and bioactive functionality within a single matrix. Unlike conventional approaches that treat these components independently, this work systematically investigates their synergistic interactions and establishes a direct correlation between composition, microstructure, and preservation performance under ambient storage conditions. This integrated approach provides new insight into the design of advanced biocomposite coatings for real-world post-harvest applications.

Accordingly, this study aims to develop and characterise PVA/CNC/ $\epsilon$ -PL biocomposite films, as well as assess their efficacy as edible coatings for increasing the shelf life of fresh chillies. Optical microscopy (OM), Fourier transform infrared spectroscopy (FTIR), Scanning electron microscopy (SEM), and X-ray diffraction (XRD) were used to analyse structural, chemical, and morphological properties, while preservation performance was measured by weight loss and visual quality after 21 days of storage. This study sheds fresh light on the development of sustainable, high-performance biocomposite coatings for active food packaging applications [5].

## 2. MATERIALS AND METHODS

### 2.1 Materials

The raw materials used in this study included powdered poly(vinyl alcohol) (PVA; 98–99 % hydrolyzed, molecular weight of 31,000–50,000, purchased from R&M Chemicals; cellulose nanocrystals (CNC) powder (grade NCV100–NAL90) supplied by CelluForce Inc. (Canada);  $\epsilon$ -

polylysine powder obtained from Jiangsu Yiming Biological Technology Co., Ltd. (China); and purified water.

## 2.2 Methods

### 2.2.1 Preparation of PVA/CNC/ $\epsilon$ -PL Solution

To begin, a 5 wt.% PVA solution was made by dissolving 5 g of PVA powder in 100 ml of distilled water and stirring continuously at 90 °C for 30 minutes until fully dissolved [6]. Simultaneously, the CNC suspension was diluted to the desired concentration by combining a 4 wt.% CNC suspension with deionised water, stirring constantly at 45 °C for 30 minutes to ensure uniform distribution. After cooling to room temperature, the homogenised PVA and CNC solution were mixed for 20 minutes to form a well-diluted and homogenous mixture. Following that,  $\epsilon$ -polylysine powder were added to the solution and agitated for another 15 minutes to ensure that all components were evenly distributed. The contents of all the materials are indicated in Table 1. To guarantee equal thickness across all samples, the PVA/CNC/ $\epsilon$ -polylysine film-forming solutions were kept at 10 ml each before being cast onto petri dishes. To allow complete water evaporation, the films were left at ambient temperature for two days. Finally, the dried biocomposite films were gently peeled from the petri dishes.

**Table 1:** The contents of PVA, CNC, PL in the nanocomposite films

Polyvinyl alcohol (PVA)	Cellulose nanocrystals (CNC)	$\epsilon$ -Polylysine ( $\epsilon$ -PL)
5wt%	4g	1g
5wt%	4g	3g
5wt%	4g	5g

### 2.2.2 Coating on Fresh Chillies by Dipping Technique

Fresh chillies were first washed thoroughly with distilled water to eliminate surface dirt and potential contaminants, then dried to remove excess surface moisture [7]. For each sample, 3 chillies were used and conducted in triplicate. The initial weight of each chili was recorded prior to coating. The chillies were then immersed in a coating solution containing polyvinyl alcohol, cellulose nanocrystals, polylysine powder to ensure uniform coverage. After dipping, the coated chillies were cured at room temperature ( $50 \pm 5\%$  RH) for 24 hours to allow proper adhesion of the coating. After drying, the coated chillies showed a homogenous and continuous film with no obvious defects like cracking, peeling, or phase separation, indicating good coating adherence. The coated chillies were subsequently stored and monitored for 21 days. In order to assess treatments that considerably increase the shelf life of fresh chillies beyond the usual 2 to 7 days ambient limit, a 21 days monitoring period serves as a common postharvest benchmark [8]. Quality parameters including visual appearance, and weight loss were evaluated to assess the effectiveness of the bio-composite coating in preserving chili quality and extending shelf life.

### 2.2.3 Solvent Casting

The solvent casting approach was used to create biocomposite films for covering fresh chilli peppers. Initially, a 5 wt.% polyvinyl alcohol (PVA) solution was made by dissolving 5 g of PVA powder in 100 ml of distilled water. The solution was heated and agitated at 90 degrees Celsius for 30 minutes, until the PVA was completely dissolved. Concurrently, a 4 wt.% cellulose nanocrystals (CNC) solution was created by diluting the CNC powder in deionised water, which was then agitated at 45 °C for 30 minutes to ensure uniform dispersion. After chilling the solutions to room temperature, the PVA and CNC solutions were combined and agitated for 20 minutes to create a uniform mixture.  $\epsilon$ -polylysine

( $\epsilon$ -PL) powder were then added to the solution, which was agitated for an additional 15 minutes to ensure complete dispersion [9].

The resulting biocomposite film-forming solutions, PVA/CNC/ $\epsilon$ -PL, were standardised at 10 mL per sample to ensure uniform thickness. A pipette was then used to transfer these solutions to petri dishes. The films were left at room temperature for two days to allow the water to completely evaporate, resulting in a uniform, dry film. Once fully dried, the films were carefully removed from the petri dishes. The solvent casting approach made it easier to create homogeneous, thin films with the appropriate thickness and characteristics for covering fresh chillies. This approach allowed for the inclusion of the biopolymer matrix with CNC, polylysine resulting in a strong covering that may improve chilli preservation.

## 2.3 Characterizations

### 2.3.1 Optical Microscopy (OM)

The surface morphology of the produced biocomposite films was characterized using an image analyzer integrated with a metallurgical microscope and high-resolution camera (camera magnification used: lens 20x - 100x magnification). Film samples were placed on clean glass slides and examined under appropriate magnification to assess surface uniformity, additive dispersion, and the presence of defects such as aggregation or phase separation.

### 2.3.2 Fourier Transform Infrared Spectroscopy (FTIR)

Fourier transform infrared spectroscopy (FTIR) was used to detect functional groups and study intermolecular interactions in biocomposite films. Spectra were obtained across a wavenumber range of 4000-400  $\text{cm}^{-1}$  using an FTIR spectrometer in attenuated total reflectance mode. The spectra were analysed to identify peaks for O-H, C-H, C=O, and amide groups, showing interactions between PVA, CNC, and  $\epsilon$ -polylysine.

### 2.3.3 Scanning Electron Microscopy (SEM)

Using a JEOL JSM-IT100 scanning electron microscope (SEM), the surface morphology and microstructural features of the biocomposite films were investigated. To improve electrical conductivity, film samples were divided into tiny specimens and sputter-coated with a thin layer of gold. To carefully examine filler dispersion, surface roughness, and the presence of phase separation or agglomerates, high-resolution micrographs were taken at a magnification of 500x and an accelerating voltage of 15 kV.

### 2.3.4 X-ray Diffraction (XRD)

X-ray diffraction (XRD) analysis was used to determine the crystalline structure of the biocomposite films. The observations were taken using an X-ray diffractometer with Cu  $K\alpha$  radiation ( $\lambda = 1.5406 \text{ \AA}$ ) over a  $2\theta$  range of  $5^\circ$ - $40^\circ$ . The diffraction patterns were analysed to identify changes in crystallinity and molecular ordering caused by CNC and  $\epsilon$ -poly-L-lysine inclusion.

### 2.3.5 Visual Inspection

The visual appearance of the biocomposite solutions and films was assessed for homogeneity, transparency, and the presence of defects such as aggregation or phase separation. Photographic images were recorded to enable qualitative comparison among different formulations.

### 2.3.6 Weight Loss

The weight loss of the chiles was measured gravimetrically during storage. Each sample's initial weight ( $W_0$ ) was recorded before coating, and subsequent weights ( $W_t$ ) were measured at specific intervals. The weight loss (%) was computed using the following equation:

$$\text{Weight loss (\%)} = [(W_0 - W_t)/W_0] \times 100. \quad (1)$$

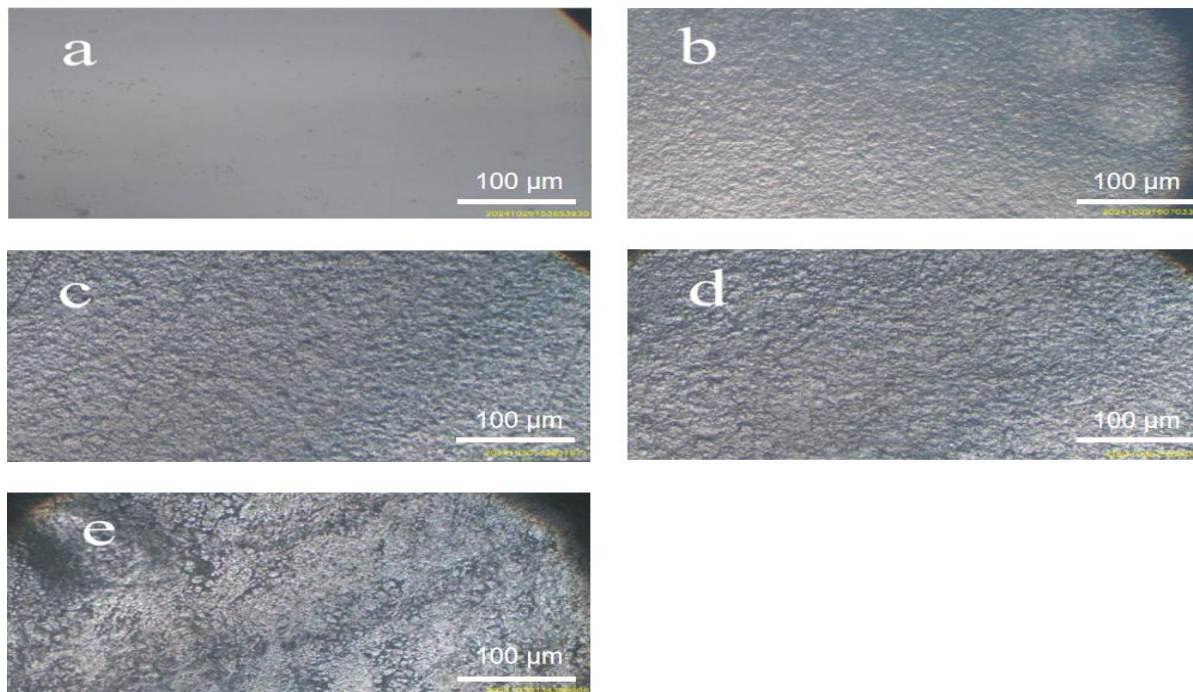
All measurements were taken in triplicate, and the average values were reported. This metric was used to assess moisture loss and the efficacy of the coatings in maintaining freshness during storage.

## 3. RESULTS AND DISCUSSION

### 3.1 Optical Microscopy (OM)

From Figure 1, the optical images reveal the surface morphology of biocomposite thin films, showing the influence of various additives and their concentrations on a polyvinyl alcohol (PVA) matrix reinforced with cellulose nanocrystals (CNC). Pure PVA film (a) exhibits a smooth and uniform surface, its structural characteristics not modified [10]. Addition of CNC (b) introduces a significant texture, indicating the formation of a composite structure that improves the mechanical and structural properties of the film.

With the incorporation of  $\epsilon$ -PL at concentrations of 1 wt%, 3 wt%, and 5 wt% (c, d, e), a progressive increase in surface roughness is observed, reflecting the interaction between  $\epsilon$ -PL and the matrix that disrupts the uniformity while potentially improving antimicrobial or functional properties. At higher  $\epsilon$ -PL concentrations, especially 5 wt%, the surface becomes highly irregular, suggesting phase separation or aggregation.



**Figure 1:** The optical images of biocomposite thin films (a) PVA; (b) PVA/ CNC; (c)PVA/ CNC/  $\epsilon$ -PL 1 wt%; (d) PVA/ CNC/  $\epsilon$ -PL 3 wt%; (e) PVA/ CNC/  $\epsilon$ -PL 5 wt%

These findings indicate a trade-off between functional benefits, such as antimicrobial activity, and film uniformity, especially at higher additive concentrations. Comparable results have been reported in the literature, where excessive additive concentrations in the polymer matrix led to aggregation and disrupted uniformity. This suggests that an optimal additive level, perhaps in the range of 1–3wt.%, is important to balance the enhanced properties with structural integrity, which is important for applications such as packaging that require both mechanical strength and antimicrobial activity.

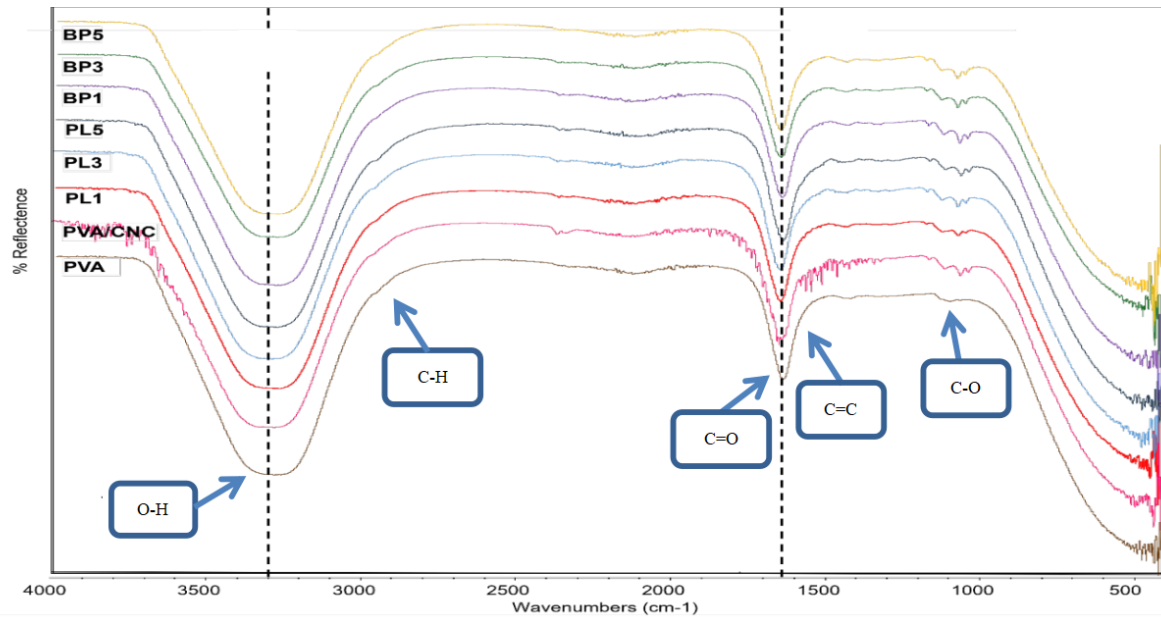
### 3.2 Fourier Transform Infrared Spectroscopy (FTIR) Analysis

The FTIR spectra of the PVA/CNC/ $\epsilon$ -polylysine ( $\epsilon$ -PL) biocomposites exhibit key functional group interactions, confirming successful incorporation of these bioactive components. A broad absorption peak between 3200–3500  $\text{cm}^{-1}$  (in Table 2), corresponding to O-H stretching, is present in all spectra, indicating strong hydrogen bonding interactions between PVA, CNC, and water molecules [11]. As it shown in Figure 2, PVA/CNC/ $\epsilon$ -PL composite, the intensity of this peak increases due to the presence of N-H stretching from polylysine. The C-H stretching vibrations (2800–3000  $\text{cm}^{-1}$ ), associated with the aliphatic chains in PVA and CNC, remain relatively unchanged, suggesting the polymer backbone remains intact. The C=O stretching peak ( $\sim$ 1650  $\text{cm}^{-1}$ ), present in all composites, corresponds to the residual acetate groups from PVA and amide bonds from polylysine. A noticeable increase in intensity in PVA/CNC/ $\epsilon$ -PL confirms the presence of amide I bonds from polylysine indicating the retention of bioactive compounds.

Further evidence of polylysine incorporation is observed in the amide II (1540  $\text{cm}^{-1}$ ) and amide III (1200–1300  $\text{cm}^{-1}$ ) peaks, which appear prominently in the PVA/CNC/ $\epsilon$ -PL spectrum. These peaks confirm the presence of protein-based functional groups, suggesting enhanced antimicrobial properties. The observed shifts and intensity variations in O-H, C=O, and amide-related peaks confirm strong molecular interactions between PVA, CNC, and bio-additives, enhancing the film's structural integrity, bioactivity, and functional properties. The presence of polylysine is expected to improve antimicrobial efficacy. These patterns are consistent with recent studies on biopolymer nanocomposites in which bioactive fillers improve functionality at the price of optical clarity [12].

**Table 2:** FTIR results of sample PVA, PVA/CNC, PVA/ CNC/  $\epsilon$ -PL 1 wt.%, PVA/ CNC/  $\epsilon$ -PL 3 wt.%, PVA/ CNC/  $\epsilon$ -PL 5 wt.% biocomposite

Bond Type	Absorption ( $\text{cm}^{-1}$ )	Functional Group
O-H stretching	3200–3500	Hydroxyl
N-H stretching	3200–3400	Amine
C-H stretching	2800–3000	Alkyl groups
C=O stretching	1650	Carbonyl from acetate or flavonoids
N-H bending	1540	Amide I
C=C stretching	1500–1600	Aromatic rings in flavonoids
C-O-C stretching	1050–1150	Glycosidic bond in cellulose
C-N stretching	1200–1300	Amide III
C-O stretching	1000–1200	Phenolic groups in BP

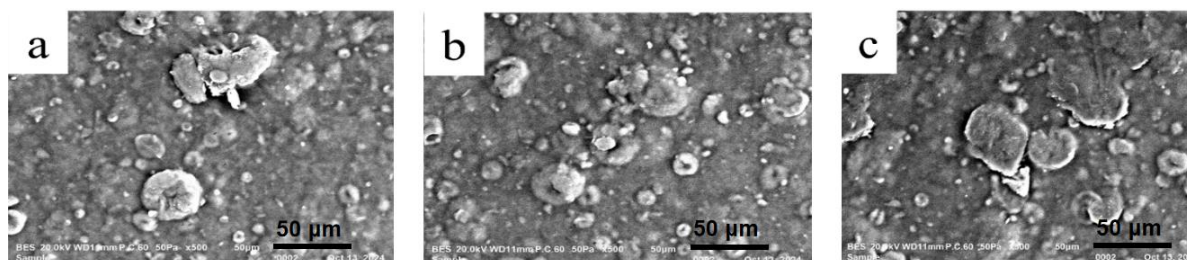


**Figure 2:** FTIR spectrum of samples PVA, PVA/CNC, PVA/ CNC/  $\epsilon$ -PL 1 wt%, PVA/ CNC/  $\epsilon$ -PL 3 wt%, PVA/ CNC/  $\epsilon$ -PL 5 wt% biocomposite

### 3.3 Scanning Electron Microscopy (SEM) Analysis

The scanning electron microscopy (SEM) images in Figure 3 provide a detailed analysis of the surface morphology of PVA/CNC-based biocomposite thin films incorporated with polylysine ( $\epsilon$ -PL) at varying concentrations (1 wt.%, 3 wt.%, and 5 wt.%). The images reveal differences in dispersion, homogeneity, and aggregation, which are critical for evaluating the structural integrity and effectiveness of these films in coating applications. SEM images of neat PVA and PVA/CNC films are not presented, as their morphology is well established in the literature, typically exhibiting smooth and homogeneous surfaces with minimal structural variation. For instance, pure PVA films have been reported to display smooth morphology without cracks or voids, while PVA/CNC composites generally exhibit well-dispersed nanocellulose within a uniform matrix [13,14]. In contrast, the incorporation of  $\epsilon$ -PL introduces significant microstructural changes; therefore, the present analysis focuses on PVA/CNC/ $\epsilon$ -PL systems to better elucidate the influence of  $\epsilon$ -PL content on film morphology. The images reveal distinct differences in dispersion, homogeneity, and aggregation, which are critical for evaluating the structural integrity and effectiveness of these films in coating applications.

For the PVA/CNC/ $\epsilon$ -PL composites, the 1 wt.% concentration in Figure 3(a) exhibits a relatively smooth surface with well-dispersed granules, suggesting good compatibility between the PVA, CNC, and polylysine. This uniformity is essential for film flexibility and stability. However, at a 3 wt.% concentration in Figure 3(b), the surface morphology becomes denser with an increased network of dispersed particles, which could enhance mechanical strength but also indicate partial aggregation. At the highest 5 wt.% concentration 3(c), the SEM image shows larger and more pronounced clusters, which may suggest poor dispersion and phase separation, potentially compromising the film's mechanical properties. Moderate concentrations (3 wt.%) provide the best balance between dispersion and structural integrity, making them more suitable for food coating applications. Similar findings have been reported in previous studies, where excessive bioactive additives in polymer matrices led to phase separation and compromised film strength. These results emphasize the importance of optimizing bio-additive concentrations to enhance the performance of biocomposite films for food preservation applications.



**Figure 3:** SEM images of biocomposite thin films (a) PVA/ CNC/  $\epsilon$ -PL 1 wt%; (b) PVA/ CNC/  $\epsilon$ -PL 3 wt%; (c) PVA/ CNC/  $\epsilon$ -PL 5 wt%

### 3.4 X-ray Diffraction (XRD) Analysis

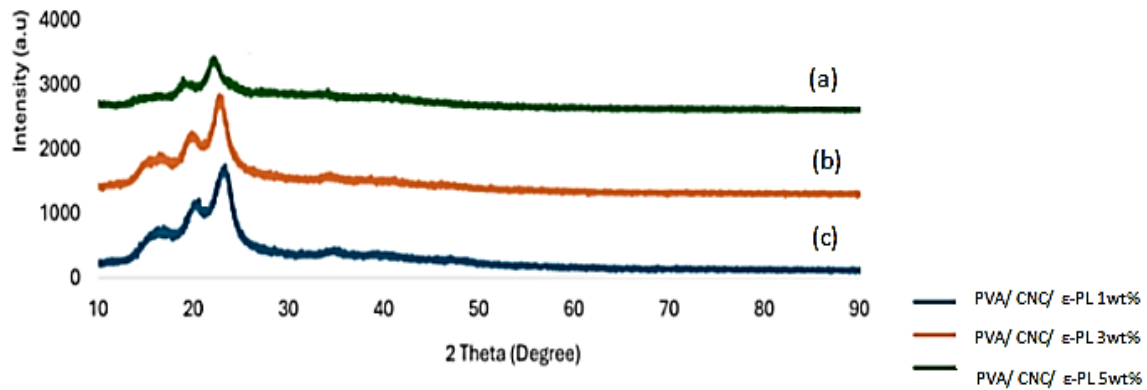
The Figure 4 shows the XRD patterns of PVA/CNC/ $\epsilon$ -PL biocomposite films reveal significant differences in crystallinity based on composition. XRD patterns of neat PVA and PVA/CNC are not presented, as their diffraction behaviour is well established in the literature, typically showing characteristic semi-crystalline peaks associated with PVA and enhanced ordering due to CNC incorporation [15]. Therefore, this study focuses on structural modifications induced by  $\epsilon$ -polylysine ( $\epsilon$ -PL), particularly changes in crystallinity and molecular ordering.

The presence of sharp peaks in the PVA/CNC/ $\epsilon$ -PL 3wt% and PVA/CNC/ $\epsilon$ -PL 5wt% spectra within the  $2\theta$  range of  $20\text{--}25^\circ$  indicates a high degree of molecular ordering and crystallinity. This finding aligns with previous research showing that CNC acts as a nucleating agent, promoting crystalline domain formation in polymer matrices. CNC acts as a reinforcing filler, increasing the crystallinity and improving the mechanical properties, such as tensile strength [16].

Samples containing PVA/CNC/ $\epsilon$ -PL 3 wt.%, and PVA/CNC/ $\epsilon$ -PL 5 wt.% exhibit moderate peak intensities, but the peaks appear broader than those. This suggests an intermediate level of crystallinity, possibly due to partial molecular ordering induced by CNC. The reinforcing effect of CNC has been widely documented, as it facilitates polymer chain alignment and enhances structural integrity. However, a noticeable reduction in crystallinity is observed in PVA/CNC/ $\epsilon$ -PL 1wt%, where peak broadening and decreased intensity within the  $2\theta$  range of  $10\text{--}15^\circ$  indicate a more amorphous structure. This suggests that polylysine dispersion within the PVA matrix disrupts crystalline domain formation, leading to a highly disordered molecular arrangement. Similar effects have been reported when bioactive compounds interfere with polymer crystallization, increasing the amorphous characteristics of the material.

Pure PVA has a semi-crystalline structure with a large diffraction peak at  $2\theta = 19\text{--}20^\circ$ , which indicates partially organised polymer chains [17]. The incorporation of cellulose nanocrystals (CNC) improves crystallinity due to their highly ordered structure and role as nucleating agents, encouraging better chain alignment in the PVA matrix. In contrast,  $\epsilon$ -poly-L-lysine ( $\epsilon$ -PL) may somewhat disrupt crystalline areas by hydrogen bonding and electrostatic interactions with PVA, which influence chain packing [18].

The  $\epsilon$ -PL interactions with CNC and PVA, which facilitate polymer chain alignment, whereas polylysine-containing samples exhibit more amorphous characteristics due to molecular dispersion. These structural variations significantly influence the mechanical, barrier, and coating properties of the biocomposite films for fresh chili coating applications.

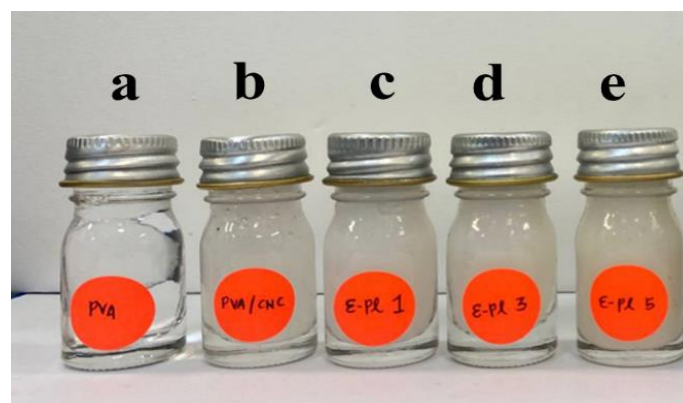


**Figure 4:** XRD results of nanocomposite films (a) PVA/ CNC/  $\epsilon$ -PL 1 wt%, (b) PVA/ CNC/  $\epsilon$ -PL 3 wt%, (c) PVA/ CNC/  $\epsilon$ -PL 5wt%

### 3.5 Visual Inspection

Figures 5 (a-h) show the visual appearance of the prepared PVA-based biocomposite solutions prior to film casting, which provides preliminary but significant information about component compatibility, dispersion, and potential intermolecular interactions. The neat PVA solution (a) is transparent and colourless, indicating full dissolution and homogeneity of the polymer chains in an aqueous media. After incorporating CNC (b), the solution becomes slightly murky, which is typically attributed to light scattering from well-dispersed nanoscale cellulose domains and the creation of hydrogen-bonded PVA-CNC networks.

The PVA/CNC/ $\epsilon$ -PL solution Figure 5(c-e) exhibit optically homogenous solutions that gradually rise in opacity as  $\epsilon$ -PL content increases from 1 to 5 wt%. The lack of apparent phase separation or sedimentation suggests good miscibility and strong electrostatic and hydrogen-bonding interactions between the cationic  $\epsilon$ -PL, hydroxyl-rich CNC, and the PVA matrix. The small increase in turbidity with larger  $\epsilon$ -PL loadings is consistent with enhanced intermolecular interactions and partial disruption of PVA crystallite formation, as observed in  $\epsilon$ -PL-modified polymer systems [19,20].

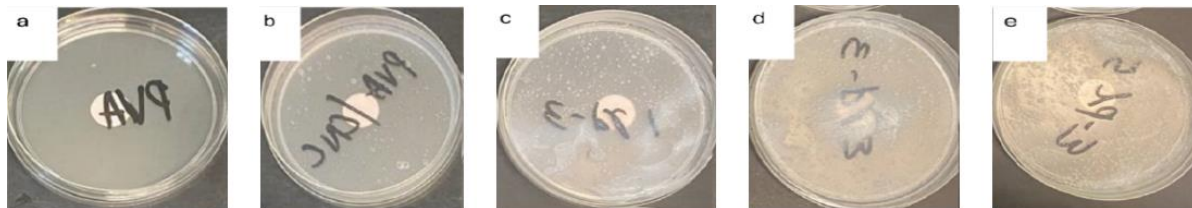


**Figure 5:** Visual inspection of bio-composite solution mixture of (a) PVA, (b) PVA/CNC, (c) PVA/ CNC/  $\epsilon$ -PL 1 wt.%, (d) PVA/ CNC/  $\epsilon$ -PL 3 wt.%, (e) PVA/ CNC/  $\epsilon$ -PL 5 wt.% (PL 5)

As shown in Figure 6, image (a) represents a PVA film, which exhibits 100 % transparency. Image (b) PVA/ CNC, depicting PVA with 5 wt.% cellulose nanocrystals (CNC), appears slightly translucent compared to pure PVA. For images (c) PVA/ CNC/  $\epsilon$ -PL 1 wt.%, (d) PVA/CNC/ $\epsilon$ -PL 3 wt.%, and (e) PVA/CNC/ $\epsilon$ -PL 5 wt.%, corresponding to PVA/CNC/ $\epsilon$ -PL with  $\epsilon$ -PL concentrations of 1 wt.%, 3

wt.%, and 5 wt.%, respectively, the films display a translucent appearance with a progressively noticeable yellow tint as the  $\epsilon$ -PL concentration increases.

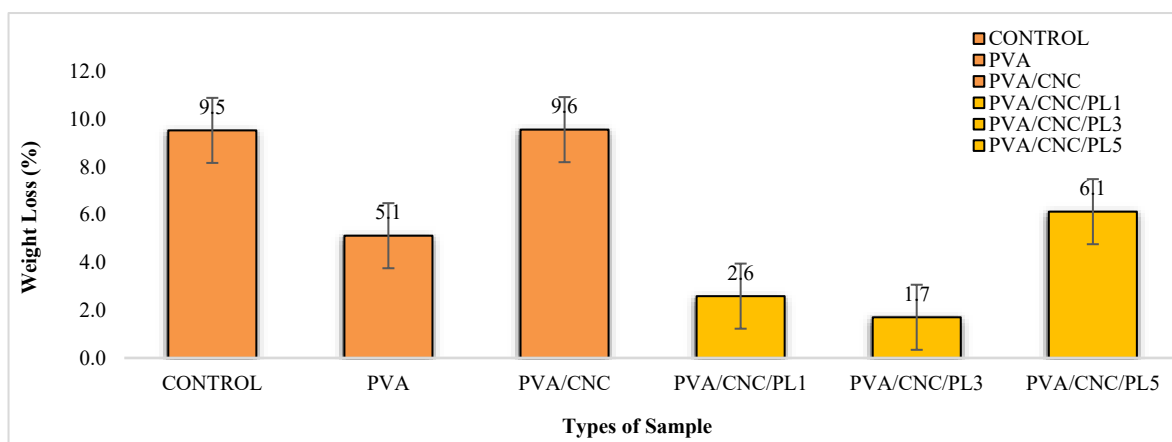
Visual inspection reveals that neat PVA film Figure 6(a) is exceedingly clear and smooth, indicating uniform chain packing. The incorporation of CNC, Figure 6(b) reduces transparency marginally while improving film uniformity, which is consistent with strong PVA-CNC hydrogen bonding [21]. The addition of  $\epsilon$ -PL, Figure 6(c-e) enhances opacity and surface heterogeneity at 3-5 wt%, indicating partial phase aggregation at higher loadings, but films remain continuous. These patterns are consistent with recent studies on biopolymer nanocomposites in which bioactive fillers improve functionality at the price of optical clarity.



**Figure 6:** Visual Inspection images of biocomposite thin films (a) PVA; (b) PVA/CNC; (c) PVA/CNC/ $\epsilon$ -PL 1 wt%; (d) PVA/CNC/ $\epsilon$ -PL 3 wt%; (e) PVA/CNC/ $\epsilon$ -PL 5 wt%

### 3.6 Weight Loss

Weight loss is a key indicator of post-harvest deterioration, primarily associated with moisture loss and respiration during storage. As shown in Figure 7, the uncoated control sample exhibited a weight loss of 9.52 % over 21 days, indicating rapid moisture evaporation in the absence of a protective barrier. The control sample consisted of uncoated chillies, which worked as a baseline for comparison to coated samples. In contrast, chillies coated with neat PVA film showed reduced weight loss (5.11 %), attributed to the formation of a semi-permeable film that restricts water vapour diffusion. The PVA/CNC-coated sample exhibited a comparable weight loss to the control (9.55 %), suggesting that CNC alone does not effectively limit moisture migration due to its hydrophilic nature. Incorporation of  $\epsilon$ -polylysine ( $\epsilon$ -PL) significantly improved coating performance. Chillies coated with PVA/CNC/ $\epsilon$ -PL at 1 wt.% and 3 wt.% showed markedly reduced weight loss of 2.58 % and 1.70 %, respectively, indicating enhanced barrier integrity and reduced transpiration. However, increasing  $\epsilon$ -PL content to 5 wt.% resulted in a higher weight loss (6.12 %), likely due to disruption of film uniformity at excessive additive concentrations [22].



**Figure 7:** Weight loss of fresh chillies coating with Control, PVA, PVA/CNC, PVA/ CNC/  $\epsilon$ -PL 1 wt%, PVA/ CNC/  $\epsilon$ -PL 3 wt%, PVA/ CNC/  $\epsilon$ -PL 5 wt% biocomposite films from Day 0 to Day 21

#### 4. CONCLUSIONS

Comprehensive characterisation of the PVA/CNC/ $\epsilon$ -PL biocomposite system demonstrates successful integration of structural reinforcement and bioactive functionality, resulting in an optimised coating for postharvest preservation. FTIR and XRD analyses revealed strong intermolecular interactions and a composition-dependent balance between CNC-induced crystallinity and  $\epsilon$ -PL-induced disruption. OM and SEM observations revealed that  $\epsilon$ -PL concentration is critical for dispersion and film uniformity, with excessive loading leading to aggregation. The solution-state behaviour also demonstrated good miscibility and colloidal stability, implying that film production was effective. The 3 wt.%  $\epsilon$ -PL formulation had the lowest weight loss (1.70 %) compared to the control (9.52 %) and neat PVA (5.11 %), demonstrating superior moisture barrier qualities. Higher  $\epsilon$ -PL concentration (5 wt.%) decreased performance due to degraded microstructural integrity, emphasising the need for compositional optimization. This study establishes a clear structure-property-performance relationship and shows that the PVA/CNC/ $\epsilon$ -PL system can be designed as a sustainable, water-processable coating with high promise for scalable application in active food packaging. However, more research into long-term storage stability, antibacterial kinetics, and migratory behaviour in real-world supply chain settings is needed to completely confirm its commercial usefulness. For the novelty, this study introduces distinctive design multifunctional biocomposite coating by synergistically combining cellulose nanocrystals and  $\epsilon$ -polylysine inside a PVA matrix to enhance mechanical strength, barrier characteristics, and antibacterial activity. The study determines the best  $\epsilon$ -PL concentration (3 wt.%) for fresh chilli postharvest preservation.

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#### Author contributions

All authors contributed toward data analysis, drafting and critically revising the paper and agree to be accountable for all aspects of the work.

#### Disclosure of conflict of interest

The authors have no disclosures to declare.

#### Compliance with ethical standards

The work is compliant with ethical standards.

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