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*Article*

# RETRACTED: Cellulose Nanocrystals/ZnO as a Bifunctional Reinforcing Nanocomposite for Poly(vinyl alcohol)/Chitosan Blend Films: Fabrication, Characterization and Properties

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**Abstract:** In this study, cellulose nanocrystals/zinc oxide (CNCs/ZnO) nanocomposites were dispersed as bifunctional nano-sized fillers into poly(vinyl alcohol) (PVA) and chitosan (Cs) blend by a solvent casting method to prepare PVA/Cs/CNCs/ZnO bio-nanocomposites films. The morphology, thermal, mechanical and UV-vis absorption properties, as well antimicrobial effects of the bio-nanocomposite films were investigated. It demonstrated that CNCs/ZnO were compatible with PVA/Cs and dispersed homogeneously in the polymer blend matrix. CNCs/ZnO improved tensile strength and modulus of PVA/Cs significantly. Tensile strength and modulus of bio-nanocomposite films increased from 55.0 to 153.2 MPa and from 395 to 932 MPa, respectively with increasing nano-sized filler amount from 0 to 5.0 wt %. The thermal stability of PVA/Cs was also enhanced at 1.0 wt % CNCs/ZnO loading. UV light can be efficiently absorbed by incorporating ZnO nanoparticles into a PVA/Cs matrix, signifying that these bio-nanocomposite films show

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good UV-shielding effects. Moreover, the biocomposites films showed antibacterial activity toward the bacterial species *Salmonella choleraesuis* and *Staphylococcus aureus*. The improved physical properties obtained by incorporating CNCs/ZnO can be useful in variety uses.

**Keywords:** biocomposites; cellulose nanocrystals; bi-functional filler; poly(vinyl alcohol)/chitosan blend; UV absorption

## 1. Introduction

The dispersion of reinforcing nano-sized particles into a continuous polymer host to form a nanocomposite has attracted great attention in recent years, because it can provide important enhancements in physical properties at very low levels of the nano-sized fillers [1]. Utilizing mechanically robust nano-sized reinforcements, for example nanoclays, carbon nanotubes, graphite and inorganic nanoparticles, into polymer hosts to enhance polymers' properties has been widely exploited [2]. Incorporation of cellulose nanocrystals as a reinforcing material for emerging novel nanocomposite products has been recently considered. Cellulose nanocrystals (CNCs) as reinforcement have some benefits over other types of nano-sized fillers, as they are inexpensive, renewable and biocompatible. These crystalline rod-like particles have been extracted after acid hydrolysis of natural cellulose derived from a diversity of renewable sources including wood, cotton, ramie, bacteria, and tunicates [3]. Most importantly, high crystalline CNCs have remarkable mechanical properties. The modulus of CNCs was determined in the range of 100–160 GPa [4]. In spite of cellulose nanocrystals' advantages, utilizing cellulose nanocrystals as alternative mono-functional fillers in polymer phases, only promotes a small number of polymers' properties [5]. This feature limits the application of nanocomposites in various fields. Incorporation of a mixture of different nano-sized fillers into polymer matrices may be desirable to fabricate nanocomposites with more and enhanced properties. Inorganic nanoparticles with excellent physical and chemical properties are important nano-sized reinforcements which have been successfully applied in the functionalization of polymer materials [6–8]. Among the various types of inorganic nanoparticles, zinc oxide nanoparticles (ZnO-NPs) has attained increased attention and is extensively utilized in a diversity of applications including functional devices, catalysts, pigments, optical materials, cosmetics, UV-absorbers, and additives in many industrial products [9–11]. Lately, the antimicrobial ability of ZnO-NPs with sizes of less than 100 nm has been reported [12]. The homogenous dispersion of inorganic nanoparticles into polymer phase has important effects on the final properties of composites. Inorganic nanoparticles tend to aggregate, owing to a large surface area and high surface energy [13,14]. In order to inhibit nanoparticles aggregation and improve dispersion during their integration into polymer matrix, one of the most effective methods is synthesizing of nanoparticles in the presence of surfactants or polymeric ligands [15]. Cellulose-based nano-materials have been widely used as templates, scaffolds, and carriers to synthesize inorganic nanoparticles [16–19]. On the other hand, CNC with abundant hydroxyl groups on its structure is compatible with water based polymer matrices. This effect helps

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to aid the homogenous dispersion of inorganic nanoparticles/CNCs nano-sized fillers blend with hydrophilic based polymers.

Poly(vinyl) alcohol/chitosan blends have attracted more attention as a biodegradable polymer blends due to their excellent biocompatibility and suitable physical properties, which can be used in environmentally friendly materials such as packaging, membrane filtration, dye adsorption and biomedical materials for controlled release, improved comfort, reduced irritation, and tissue engineering [20–22]. The improvement of PVA/Cs's mechanical, thermal, UV shielding and antimicrobial properties is valuable.

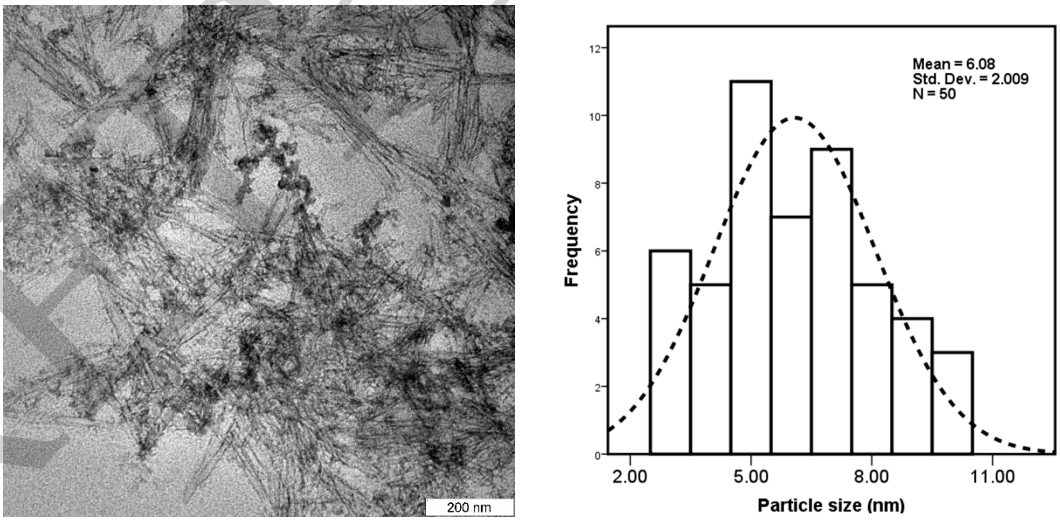
This study investigates the effect of CNCs/ZnO bi-functional nano-sized filler on the functional properties of PVA/Cs matrices through a solution casting method. Considering the functional properties of CNCs and ZnO-NPs, the incorporation of CNCs/ZnO is expected to improve mechanical, thermal properties, UV absorption and antibacterial activity of the polymer blend nanocomposites.

## 2. Results and Discussion

### *2.1. Characterization of CNCs/ZnO*

A TEM image of CNCs/ZnO deposited from a dilute aqueous suspension is shown in Figure 1. From the TEM image it can be observed that the ZnO nanoparticles with a narrow size distribution are stabilized by rod-like shapes of CNCs. The individual particles have diameters in the range of 6–10 nm with a mean size of 6.08 ± 2.00 nm.

**Figure 1.** (**a**) TEM image and (**b**) particle size distribution graph of cellulose nanocrystals/zinc oxide (CNCs/ZnO).




(**a**) (**b**)

### *2.2. Characterization of PVA/Cs-Based Films*

#### 2.2.1. X-ray Diffraction (XRD)

The X-ray diffraction patterns for CNCs/ZnO powder, PVA/Cs, and PVA/Cs/CNC/ZnO films are illustrated in Figure 2. The CNCs/ZnO shows two sets of diffraction peaks corresponding to CNCs and

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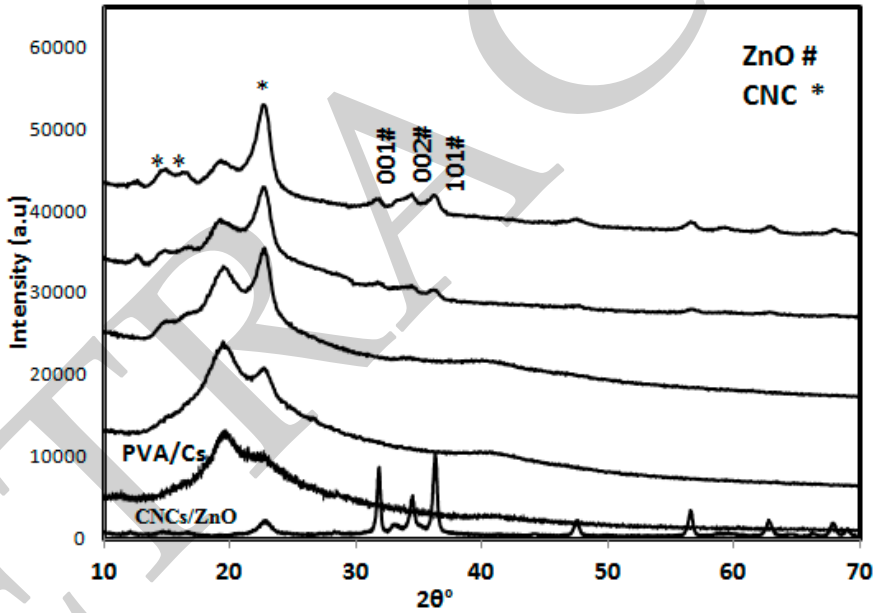
ZnO, in the 2θ range of 10°–80°. The PVA/Cs film shows the characteristic pattern of an amorphous phase with the main halo of the typical peak at a 2θ of 19.05° and another of low intensity at a 2θ of 22.15° [23]. The X-ray diffraction pattern shows the peak of PVA/Cs and several sharp and increased diffraction peaks for PVA/Cs/CNCs/ZnO bio-nanocomposite films with a relatively high CNCs/ZnO content. These peaks in the 2θ range of 10°–80° were almost the same as those of the CNCs and ZnO, though the sharp diffraction peaks increased with increasing CNCs/ZnO content. When a high level of CNC/ZnO was used, the presence of a larger number or self-agglomerated nanoparticles, caused the crystalline character, attributed to the CNC and ZnO in the bio-nanocomposite, to become clearer. There was neither a new peak nor a peak shift compared with the pure PVA/Cs, indicating that the PVA/Cs/CNCs/ZnO biocomposite films all consisted of two phase structures, *i.e.*, polymer and nanoparticles. These observations showed that the addition of the CNCs/ZnO caused an increase in the overall crystallinity of PVA/Cs polymer blend, which can be effective on the final proprieties of bio-composites films.

**Figure 2.** XRD pattern of CNCs/ZnO, poly(vinyl alcohol) and chitosan (PVA/Cs) blend and its CNCs/ZnO biocomposites.




2.2.2. Surface Morphology

The dispersion status of CNCs/ZnO nano-sized fillers in the polymer matrix was observed using FESEM; Figure 3a,c,d shows the upper surface and Figure 3b,e shows the cross section surface images of PVA/Cs blend and PVA/Cs/CNCs/ZnO 5.0 wt % biocomposites, respectively. Figure 3a shows relatively a good adhesion between polymer phases with some small pores. The addition of CNCs/ZnO caused the surface morphology of PVA/Cs blend film became rough. Figure 3d clearly indicates the morphologies related to ZnO nanoparticles and CNCs. The swell sphere and rod-like shapes are ZnO and CNCs, respectively. For details, as shown in Figure 3c, a homogeneous dispersion of the CNCs/ZnO inserted in the PVA/Cs matrix was seen, suggesting that there exists good adhesion between nano-sized fillers and matrix. Such an even and uniform distribution of the nano-sized

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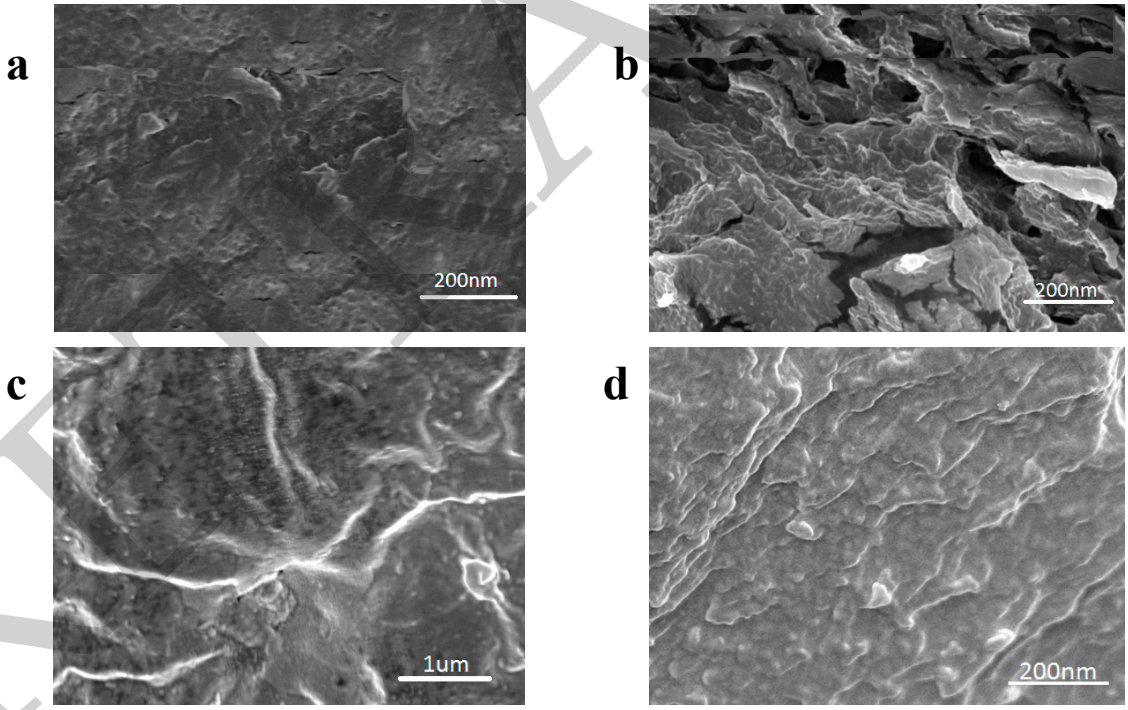
fillers in the matrix played an essential role in enhancing the mechanical performance. In general, the Zn–O–Zn bonds that form in nanoparticles in the presence of molecular water result in hard agglomerates, which obstruct the application of ZnO nanoparticles [11,24]. Considering the homogenous dispersion of CNCs/ZnO in polymer blend matrix, it can be concluded that the stabilization of ZnO nanoparticles by cellulose nanocrystals could help to increase their dispersion in the polymer blend matrix and prevents from more agglomerations. Cross section surface images (Figure 3b,e) evidently show that the addition of CNCs/ZnO has a definite effect on the adhesion of polymer phases. The homogenous dispersion of filler and good afinity between filler-matrices led to decreasing density of crack deflection sites and improved misibility of polymer phases.

The presence of elements in the biocomposite was analyzed by EDX spectrum and is presented in Figure 3e. It exhibits peaks at 0.24, 0.52, 1.0 and 2.4 keV corresponding to carbon, oxygen, Zn and sulfur respectively. The presence of cellulose nanocrystals, as they have a similar composition as the matrix, was confirmed with a weak sulfur peak which most likely originates from the sulfate group of the sulfuric acid that was used for the extraction process. Therefore EDX analysis clearly shows the presence of C, O, S and Zn which indicates that CNCs and ZnO polymers are present in the biocomposite.

**Figure 3.** FESEM micrographs of upper surfaces (**a**,**c**,**d**) and cross section surfaces (**b**,**e**) of PVA/Cs and PVA/Cs/CNCs/ZnO 5.0 wt % bio-nanocomposite, respectively; EDX spectrum (**f**) of bio-nanocomposite film.

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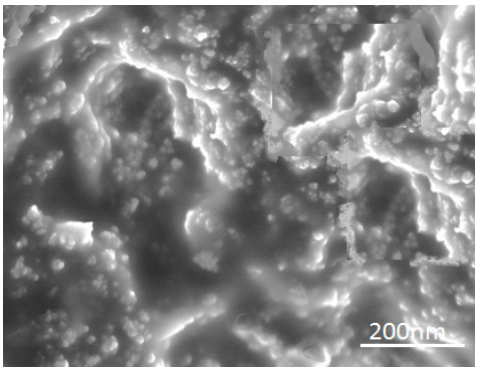
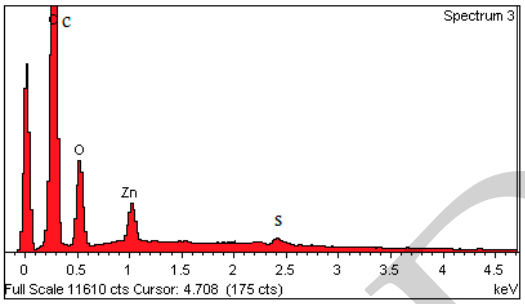



**f**




2.2.3. Mechanical Properties

The effect of the CNCs/ZnO contents on the tensile strength and tensile modulus of the PVA/Cs bio-nanocomposite films is given in Table 1. It is obvious that the addition of CNCs/ZnO significantly increased the tensile strength and modulus from 0 to 5.0 wt %, and then dropped. Tensile strength (*T*s) increased from 55.1 to 153.3 MPa, which represents a ~178% enhancement, and tensile modulus (*T*m) improved from 395 to 932 MPa, which gives a ~135% increase. This result indicates that the addition of CNCs/ZnO into the PVA/Cs blend matrix results in strong interactions between the matrix and filler; thus, restricting the matrix motion and promotes rigidity [25]. Further addition of CNCs/ZnO decreased tensile strength and modulus, but remained greater than the original PVA/Cs film. This reduction is due to inter-particle interactions which lead to the creation of weak points. On the other hand, the elongation at break gradually decreased from 0 to 7.0 wt % of filler loading with a maximum reduction at 5.0 wt %. The elongation at breaks (*E*b) decreased from 10.3 to 3.2 which shows a ~67% reduction. This phenomenon can be explained by the fact that the strong filler–matrix interaction occurred because the filler content is 5.0 wt %.

**Table 1.** Tensile data of PVA/Cs based films.

| Sample | CNCs/ZnO (wt %) | *T*s (MPa) | *T*m (MPa) | *E*b (mm) |
|---|---|---|---|---|
| PVA/Cs | 0 | 55 ± 1.6 | 395 ± 23 | 10.2 ± 0.2 |
| PVA/Cs/CNCs/ZnO | 1 | 86 ± 1.3 | 691 ± 40 | 6.3 ± 0.9 |
| PVA/Cs/CNCs/ZnO | 3 | 120 ± 2.1 | 825 ± 38 | 5.5 ± 0.4 |
| PVA/Cs/CNCs/ZnO | 5 | 153 ± 1.8 | 932 ± 36 | 3.2 ± 0.7 |
| PVA/Cs/CNCs/ZnO | 7 | 112 ± 2.7 | 801 ± 30 | 4.8 ± 0.3 |

2.2.4. Thermogravimetric Analysis

TG and DTG curves of pure PVA/Cs and PVA/Cs/CNCs/ZnO films are shown in Figure 4a,b, respectively. Thermal parameters including the onset temperature, $T_{onset}$, and the mid-point of the degradation, $T_{0.5}$, are summarized in Table 2. It can be seen that the PVA/Cs blend decomposes in two steps. According to the DTG curves, all biocomposites show two main degradation steps of PVA/Cs blend. As is evident, incorporation of CNCs/ZnO has no effect on the decomposition stages of biocomposites. However, the degradation temperature alters, which indicates the thermal stability of the biocomposites changes. It can be seen that, apart from the PVA/Cs/CNCs/ZnO 1.0 wt % sample,

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the $T_{onset}$ and $T_{0.5}$ of biocomposites decreased compared to that of the polymer blend. This result implies that the biocomposites decompose at lower temperatures than that of the original PVA/Cs matrix; *i.e.*, incorporating CNCs/ZnO into the PVA/Cs decreases the thermal stability. The change of the thermal stability of the biocomposites can be explained as follows: On the one hand, the presence of ZnO and CNCs in the PVA/Cs matrix due to interaction with polymer matrix could restrict the motion of the polymer chains or act as a thermal insulator and barrier to the volatile products formed throughout degradation, therefore this leads to a delay in thermal decomposition. On the other hand, ZnO as a semiconductor is able to generate free oxygen and oxygen vacancies in the lattice structure induced by thermal stimulation. The oxygen vacancies can absorb electrons to make active catalytical positions in ZnO, and free oxygen raises the creation of peroxy radicals to break the polymer chains. Therefore, formation of free oxygen and oxygen vacancies plays a main role in decomposing polymers [26]. The above two effects contend with each other and the thermal catalysis effect of ZnO nanoparticles overcomes most biocomposites. Consequently, the $T_{0.5}$ was shifted by 13, 15 and 19 °C towards lower temperatures when the CNCs/ZnO contents were 3.0, 5.0 and 7.0 wt % respectively. On the other hand, in comparison with these biocomposites, the $T_{0.5}$ shifted by 34 °C towards a higher temperature, when the CNCs/ZnO content was 1.0 wt %, due to the effect of heat-resistance of ZnO-NPs and interaction between filler–matrix.

**Figure 4.** TGA (**a**) and DTG (**b**) thermograms of PVA/Cs-based films.

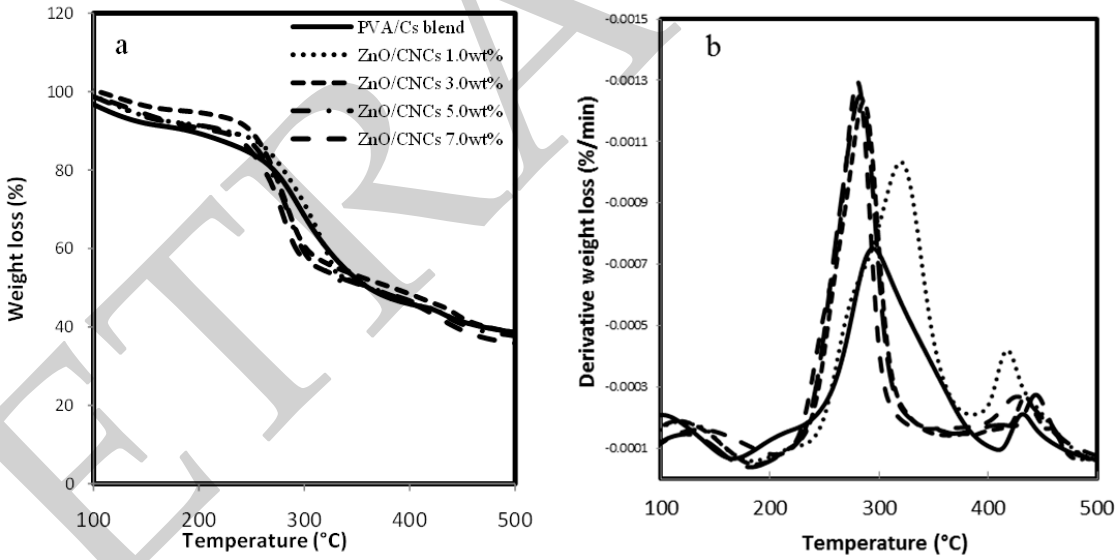


**Table 2.** Thermal data of PVA/Cs based films.

| Sample | CNCs/ZnO (wt %) | First Step | |
|---|---|---|---|
| | | $T_{onset}$ (°C) | $T_{0.5}$ (°C) |
| PVA/Cs | 0 | 217 | 294 |
| PVA/Cs/CNCs/ZnO | 1 | 215 | 328 |
| PVA/Cs/CNCs/ZnO | 3 | 212 | 281 |
| PVA/Cs/ CNCs/ZnO | 5 | 211 | 279 |
| PVA/Cs/CNCs/ZnO | 7 | 209 | 275 |

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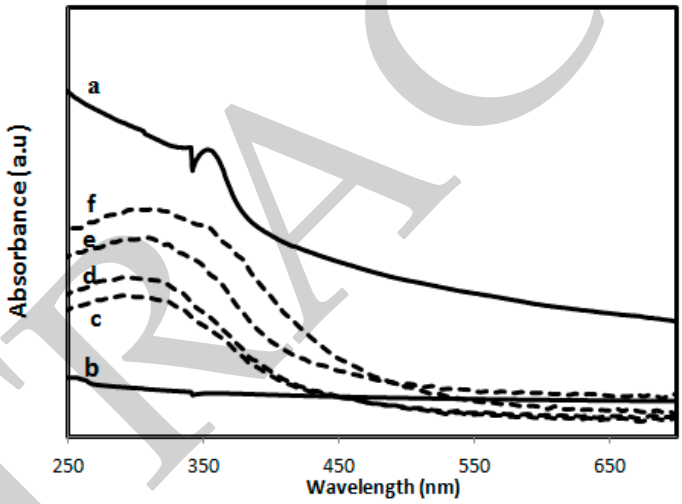
2.2.5. Ultraviolet Absorbance

The optical properties of CNCs/ZnO and PVA/Cs/CNCs/ZnO biocomposites observed by ultraviolet-visible absorption spectrophotometry are shown in Figure 5. In contrast to PVA/Cs blend, all biocomposite films show an absorption band at about 320 nm which is attributed to the band gap of the ZnO-NPs [27], and this is clearly evident for CNCs/ZnO at about 350 nm. The absorption peak of PVA/Cs/CNCs/ZnO samples showed a noticeable blue-shift phenomenon, indicating strong interactions between matrix and ZnO-NPs. Such shifting of the absorption bands owing to the interaction between a polymer matrix and ZnO has been reported in the literature [28,29]. With increasing ZnO content, both the Uv absorbance and the intensity of the absorption peaks increased. Maximum absorption was obtained for the polymer blend with a filler loading of 7.0 wt %. The PVA/Cs/CNCs/ZnO biocomposites may successfully protect against ultraviolet light, and could potentially be used as ultraviolet-shielding materials.

**Figure 5.** Ultraviolet spectra of CNCs/ZnO (a); PVA/Cs blend (b); and PVA/Cs/CNCs/ZnO biocomposites from 1.0 to 7.0 wt % filler loading (c–f).




2.2.6. Zinc Release from PVA/Cs/CNCs/ZnO Bio-Nanocomposite

In order to understand the resultant toxicology of bio-nanocomposites, the amount of $Zn^{2+}$ released from PVA/Cs/CNCs/ZnO samples was measured in water using the Inductively Coupled Plasma (ICP) technique. The application of such composites in wound healing might cause some health threats if the nanoparticles or free ions are released in high amounts [29]. As shown in Figure 6, very small amounts of $Zn^{2+}$ released from the films during the complete testing period. The concentration of $Zn^{2+}$ released increased slowly with time and reached to 0.7 (mg/L) after day 8 incubation for PVA/Cs/CNCs/ZnO 7.0 wt %. This slow release indicates the strong interaction of ZnO with PVA/Cs matrix [29].

**Figure 6.** The amount of zinc released from PVA/Cs/CNCs/ZnO bio-nanocomposite films.

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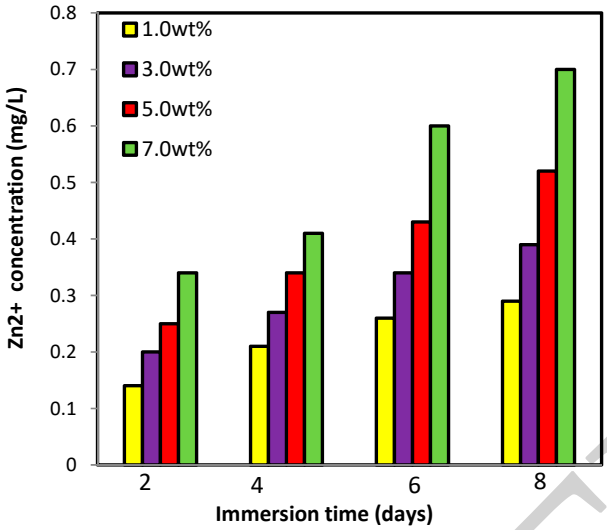


2.2.7. Antibacterial Assessment

The photocatalytic antimicrobial power of the biocomposite films was evaluated by measuring the inhibition zones of *S. aureus* and *S. choleraesuis* cells under ultraviolet light, as shown in Figure 7. Table 3 shows the average inhibition zones for all the biocomposite samples, in comparison with the pure PVA/Cs film. In contrast with PVA/Cs film, the PVA/Cs/CNC/ZnO-biocomposites show an inhibition zone against the bacteria. The antibacterial activity of the biocomposites can be explained by photocatalysis, zinc release and accumulation of ZnO-NPs [30]. When ZnO nanoparticles are under light irradiation, electron-hole pairs are generated. The holes split $H_2O$ molecules from ZnO into $OH^-$ and $H^+$. The hole ($h^+$) reacts with $OH^-$ on the surface of the nanoparticles, generating hydroxyl radicals ($OH^+$), superoxide anion ($O^{2-}$) radicals, and perhydroxyl radicals ($HO_2^+$). These highly active free radicals harm bacterial cells, resulting in their decomposition and destruction [31]. On the other hand; the release of Zn ions may bind to proteins to deactivate them, interact with microbial cell envelope to cause structural change and permeability, and interact with microbial nucleic acids to inhibit microbial replication. In addition, accumulation of ZnO nanoparticles in the microbial membrane could result in membrane disorganization and microbial cellular internalisation [32]. As has been shown, the strong interaction of ZnO with matrix reduces the effect of the two last parameters, thus it can be concluded that the antimicrobial ability of biocomposites is mainly due to photocatalysis effect of ZnO nanoparticles. As given in Table 3, PVA/Cs/CNC/ZnO 7.0 wt % showed strong antimicrobial effects against bacteria especially towards *S. aureus* microbial cells. The difference in antibacterial activity of the biocomposites against these two types of bacteria can be related to structural and chemical compositional differences in the cell membrane.

**Figure 7.** Inhibition zone images of PVA/Cs/CNCs/ZnO biocomposites against (**a**) Gram-positive and (**b**) Gram-negative bacteria.

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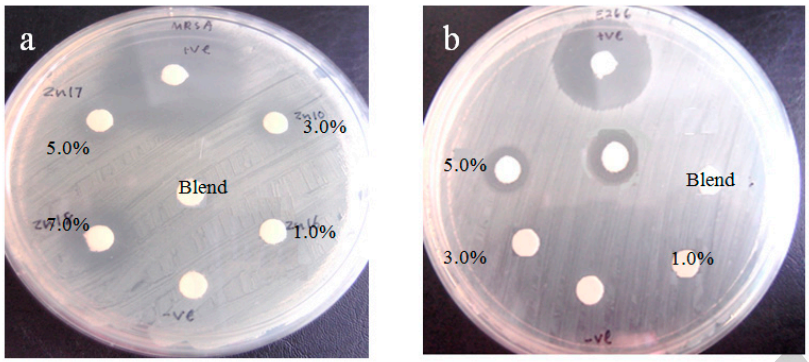


**Table 3.** Inhibition zone data of PVA/Cs-based film.

| Filler Content wt % | Diameter of Zone (mm) | |
|---|---|---|
| | Gram-Positive | Gram-Negative |
| 0 | - | - |
| 1.0 | - | - |
| 3.0 | 5.4 | - |
| 5.0 | 6.3 | 4.9 |
| 7.0 | 7.1 | 5.4 |

## 3. Experimental Section

### *3.1. Materials*

All the chemicals were analytical grade and used as received without more purification. Cotton cellulose from filter paper (Q1, Whatman) was supplied by Fisher Scientific (Pittsburgh, PA, USA). Sulfuric acid (95%–98%, reagent grade) was purchased from Scharlau (Barcelona, Spain). Ethanol and sodium hydroxide were provided by Sigma Aldrich (St. Louis, MO, USA). Zinc acetate dehydrate (99%) used as precursor, was provided by Merck (Darmstadt, Germany). Chitosan with a 85% degree of deacetylation and "345,500" $g \cdot mol^{-1}$ average molecular weight was obtained by the Kuala lumpur, Malaysian Nuclear Agency. PVA with a 99% of degree of hydrolysis and average molecular weight of "89,000" $g \cdot mol^{-1}$ was purchased from Sigma Aldrich (St. Louis, MO, USA).

### *3.2. Preparation of CNCs/ZnO Bifunctional Nano-Sized Filler*

The CNCs/ZnO was synthesized according to our previous work [33]. Firstly, the CNCs were suspended in deionized water, then mixing with zinc acetate dehydrate ($Zn(Ac)_2 \cdot 2H_2O$) in ethanol by magnetic stirring. The weight ratios of Zn:CNCs was 1:2. After complete mixing, a sodium hydroxide solution (5 mol/L) was added drop-wise to the mixed solutions with gentle stirring at 80 °C. The CNCs/ZnO nanocomposites were separated into different phases from the suspension by centrifugation and were then washed using distilled water to remove the by-products and extra CNCs. After complete washing, the samples were dried at 120 °C for 1 h for complete transformation of the remaining zinc hydroxide to zinc oxide.

### *3.3. Preparation of PVA/Cs/CNCs/ZnO Biocomposite Films*

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The PVA/Cs blend was prepared as a described work [34]. Briefly, specific quantity of PVA (5.0 *w*/*v* %) was added into the 1.0 *w*/*v* % chitosan solution to obtain PVA/Cs molar ratio of (3:1). Then CNCs/ZnO were dispersed in a solution of distilled water (100 mL) and ultrasonicated for 30 min. The CNCs/ZnO nano-sized filler loading level (0, 1, 3, 5 or 7 wt %) was based on the amount of PVA/Cs blend. The above suspensions were added into 100 mL of the PVA/Cs under strong string for 2 h at 70 °C. The mixtures were casted in glass petri dishes and then dried in ambient temperature to obtain the composite films. The dried composite films were roasted at 45 °C for 6 h. Finally, a series of nanocomposite films with a thickness of ~0.2 mm were prepared.

### *3.4. Characterization*

The X-ray analysis was carried out using Philips X'pert PXRD (Almelo, The Netherlands). The X-ray beam was nickel-filtered Cu (λ = 1.542 A°) and the instrument was operated at 40 kV and 30 mA. The scanning scope of 2θ was 5°–80° at room temperature. The morphology and components of the biocomposite film were performed using field emission scanning electron microscope (FESEM) and energy-dispersive X-ray (EDX) respectively, (JSM-6360LA Philips, Eindhoven, The Netherlands). The specimens were mounted on a metal stub using carbon tape and then gold coated using a sputter coater. The size and morphology of the CNCs/ZnO were seen using a Hitachi H-700 transmission electron microscope (Tokyo, Japan) with an acceleration voltage of 120 kV at room temperature. The TEM sample was prepared via dropping the sample suspension on a Cu grid coated with carbon film, and then the specimens were negatively stained with 1% uranyl acetate and allowed to dry at room temperature. The tensile properties of the original polymer blend and bio-nanocomposites were measured as per ASTM D 638 test methods, using an Instron 4032 Universal Testing Machine (Instron, Pfungstadt, Germany). The samples, cut into dumb-bell shapes, employed a steel template and router machine. Seven specimens were tested and the average result of the values of five specimens was taken. The thermal behavior of the samples was recorded with a thermogravimetric analyzer TGA7 (Perkin-Elmer, Waltham, MA, USA) in a nitrogen atmosphere at a heating rate of 10 °C/min from 25 to 700 °C. The optical properties of the samples were investigated by using a UV-vis-NIR Spectrophotometer (Lambda 25-Perkin Elmer, Waltham, MA, USA). The samples were placed in chamber and then scanned in the range of 200–800 nm wavelengths at room temperature.

### *3.5. Zinc Release*

The quantity of zinc ($Zn^{2+}$) released from each sample was evaluated by dipping the dried PVA/Cs/CNCs/ZnO films (2 × 2 $cm^2$) in 5 mL deionized water for various times, after that the residual concentration of $Zn^{2+}$ in the water was determined using Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES) (model Perkin Elmer 1000,Waltham, MA, USA).

### *3.6. Antibacterial Measurements*

The samples were evaluated for antibacterial activity against Gram-negative *Salmonella choleraesuis* and Gram-positive *Staphylococcus aureus*. The bacterial grown overnight in nutrient broth. The bacterial inoculum was standardized to 0.5 MF units, which meant that approximately $10^8$ colony-forming

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units of each bacterium were inoculated on a plate. The samples with diameter of 3 mm, were placed onto an plate that was inoculated with bacteria. The plates were inverted and incubated at 37 °C under UV light for 24 h, following which the zone of complete inhibition was measured. Three replicate tests were carried out in the same conditions for the each sample. The reported inhibition zone values represent the average of all three samples.

## 4. Conclusions

The reinforced composites of poly(vinyl alcohol)/chitosan matrix with cellulose nanocrystals/ZnO were prepared and the potentiality of bifunctional nano-sized fillers was measured on the structure and properties of resulting hybrids. Mechanical, thermal, Uv-vis and morphological properties indicated, in general, promising matrix–filler interactions arising from effective dispersion of CNCs/ZnO in a polymer blend matrix. CNCs/ZnO significantly increases tensile strength and the modulus of PVA/Cs-based films to an optimum value (5.0 wt %), and then slowly decreases. Thermal resistance enhanced only at 1.0 wt % filler loading. Nano-ZnO impregnation also improved the absorbance of UV radiation in comparison to pure PVA/Cs. The biocomposites were effective against *S.choleraesuis* and *S. aureus*, with dependence on the amount of the CNCs/ZnO added. These biocomposites have potential applications in medical, packaging, and UV-shielding materials.

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## Author Contributions

Susan Azizi: She performed experiments, characterized, analyzed data and wrote the paper.

Mansor Ahmad: He designed and supervised experiments, analyzed data and monitoring the writing the paper.

Nor Azowa Ibrahim: She designed and analyzed data.

Mohd Zobir Hussein: He analyzed data and edited the paper.

Farideh Namvar: She performed some experiments and characterization.

## Conflicts of Interest

The authors declare no conflict of interest.

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