



# Green-synthesized silver nanoparticle coating on paper for antibacterial and antiviral applications

Natwat Srikhao<sup>1</sup> · Artjima Ounkaew<sup>1</sup> · Natnaree Srichiangsa<sup>1</sup> ·  
Supranee Phanthanawiboon<sup>2</sup> · Thidarut Boonmars<sup>3</sup> · Atchara Artchayasawat<sup>3</sup> ·  
Somnuk Theerakulpisut<sup>4</sup> · Manunya Okhawilai<sup>5,6</sup> · Pornnapa Kasemsiri<sup>1</sup>

Received: 15 March 2022 / Revised: 9 September 2022 / Accepted: 10 October 2022

© The Author(s), under exclusive licence to Springer-Verlag GmbH Germany, part of Springer Nature 2022

## Abstract

The use of active packaging has attracted considerable attention over recent years to prevent and decrease the risk of bacterial and viral infection. Thus, this work aims to develop active packaging using a paper coated with green-synthesized silver nanoparticles (AgNPs). Effects of different silver nitrate (AgNO<sub>3</sub>) concentrations, viz. 50, 100, 150, and 200 mM (AgNPs-50, AgNPs-100, AgNPs-150, and AgNPs-200, respectively), on green synthesis of AgNPs and coated paper properties were investigated. A bio-reducing agent from mangosteen peel extract (ex-Garcinia mangostana (GM)) and citric acid as a crosslinking agent for a starch/polyvinyl alcohol matrix were also used in the synthetic process. The presence of AgNPs, ex-GM, and citric acid indicated the required synergistic antibacterial activities for gram-positive and gram-negative bacteria. The paper coated with AgNPs-150 showed complete inactivation of virus within 1 min. Water resistance and tensile strength of paper improved when being coated with AgNPs-150. The tensile strength of the coated paper was found to be in the same range as that of a common packaging paper. Result revealed that the obtained paper coated with AgNPs was proven to be effective in antibacterial and antiviral activities; hence, it could be used as an active packaging material for items that require manual handling by a number of people.

**Keywords** Silver nanoparticles · Green synthesis · Active packaging · Antibacterial activities · Antiviral activity

---

✉ Pornnapa Kasemsiri  
pornkas@kku.ac.th

Extended author information available on the last page of the article

## Introduction

Paper is a cellulosic material, which has been used for versatile purposes such as packaging, printing/writing, household products, filtration, and healthcare items because of its biodegradability, light weight, reusability, and recyclability [1, 2]. In improving and uncovering new functions to paper, a common and simple technique such as coating has been applied to achieve new properties [3, 4]. Substances such as bioactive agents, wax, and biopolymers have been used for paper coating to extend its applications [5–7].

The use of nanotechnology to create nanomaterials has been quite common in coating applications because of the high surface area of nanoparticles with regard to their volume, low utilization of chemical agents, and less consumption of energy [8, 9]. Among available nanomaterials, silver nanoparticles (AgNPs) are widely used because of their outstanding properties such as antimicrobial activities [10, 11] and less toxicity for mammalian cells [12, 13]. Coating of AgNPs on various substrates has been used in personal care products [2], food packaging [14, 15], and medical instruments [16]. The preparation of AgNPs can be achieved by various techniques. Among the techniques, the green synthesis of AgNPs is more ecofriendly than other methods because of the use of materials with low toxicity [17–19]. Several bio-based compounds such as terpenoids, glycosides, alkaloids, and phenolics can be used as bio-reducing agents [10, 20]. Recently, the new trend of using bio-based extracts from agricultural [21, 22], food, and beverage wastes [23] has attracted considerable attention. Ounkaew et al. [7] prepared the paper coated with green-synthesized AgNPs using spent green tea. They found that the coating of AgNPs on paper enhanced water resistance and inhibited the growth of *Staphylococcus aureus* (*S. aureus*) and *Escherichia coli* (*E. coli*). The presence of extracted spent green tea in AgNP synthesis could reduce the reaction time from 2 h to 20 min. Panzella et al. [23] proposed the green synthesis of AgNPs using spent coffee grounds (SCG). The obtained phenolic compounds in SCG such as chlorogenic acid and caffeic acid served as bio-reducing agents. The SCG containing 0.3%wt of AgNPs could inhibit the growth of *Pseudomonas aeruginosa* (*P. aeruginosa*), *E. coli*, and *S. aureus*. Ounkaew et al. [22] observed that the use of agricultural waste such as sugarcane leaf combined with 20 wt% citric acid for in situ green synthesis of AgNP nanocomposite films provided a synergistic effect of antibacterial activities. In addition, the presence of citric acid as a crosslinker in nanocomposite films based on starch and polyvinyl alcohol (PVA) created ester linkages, which led to an increase in mechanical properties and water resistance. Apart from the use of wastes from agriculture and beverage as bio-reducing agents, the waste from fruit peels would be an attractive alternative source because of the availability of enriched phenolic compounds [24, 25]. *Garcinia mangostana* (GM) or purple mangosteen peel consists of phenolic compounds such as xanthenes, which can be applied in medical applications [26]. Lee et al. [27] observed that the use of GM peel for AgNP synthesis in an acidic condition with pH of about 4 yielded nanoparticles in spherical shapes and a uniform dispersion of particles. Green-synthesized AgNPs using

GM peel could be applied as an anticancer drug nanocarrier. Although a number of studies have focused on green synthesis of AgNPs for various applications, particularly antibacterial activities, little information is found on antiviral activities and other properties. At present, considering that microbial pathogens such as viruses are also a major public health concern [28], the development of coating materials for the inhibition of microorganism growth could be a new and effective approach to prevent bacterial and viral infection [29, 30].

In this study, the coating of AgNPs on paper was established by using extracted GM peel as a bio-reducing agent and starch/PVA copolymer as a binder. Different concentrations of silver nitrate for the green synthesis of AgNPs were studied. Moreover, antibacterial and antiviral activities of paper coated with AgNPs, water resistance, and tensile strength were investigated.

## Experimental

### Materials

Cassava starch with 17% amylose and 12.17% moisture was purchased from Bangkok Interfood Ltd., Thailand; PVA with an average molecular weight of 1700–1800 was purchased from Laboratory Reagents & Fine Chemical. Citric acid with purity of 99.5% was purchased from RCI Labscan Limited. GM peel powder with a particle size of 180–250  $\mu\text{m}$  was supplied from local shops in Khon Kaen, Thailand.

### Preparation and characterization of ex-GM

The ex-GM was prepared by using the ratio of GM peel powder to deionized water at 1:60. The mixture of GM peel powder and deionized water was heated at 90 °C for 5 min with constant agitation. Then, the mixture was filtered through a qualitative filter (no. 1) to obtain ex-GM sample. The total phenolic content (TPC) of ex-GM was determined using the Folin–Ciocalteu method and gallic acid as standard according to Tronchuen et al. [31]. A UV–vis spectrophotometer (Agilent Cary 60 UV–Vis Spectrophotometer) was used in the analysis to obtain TPC. Ex-GM had a TPC of  $37.48 \pm 1.06$  mg GAE/g of GM peel powder.

### Preparation of paper coated with AgNPs

First, the solutions of cassava starch and PVA were prepared separately. 2.5 g of cassava starch was dissolved in 50 mL of ex-GM by using a magnetic stirrer for 20 min at 90 °C. 2.5 g of PVA was also dissolved in ex-GM at 95 °C for 60 min. Afterward, the solutions of starch and PVA were mixed for another 5 min at 90 °C. Silver nitrate solutions with concentrations of 0, 50, 100, 150, and 200 mM were added into the starch/PVA solution and stirred for 12 h at 90 °C. The obtained solution was cooled down to room temperature, and then 5 g of citric acid was added and stirred for another 5 min. The homogenous mixture solution was cast on a piece of filter paper

using a doctor blade with a gap of 200  $\mu\text{m}$  to evenly spread the mixture solution. The coated paper was then dried at 50  $^{\circ}\text{C}$  for 30 min.

### Characterization of paper coated with AgNPs

A UV–Vis spectrophotometer (Perkin-Elmer, Lambda 35, spectrometer) was used to characterize the green-synthesized AgNPs at 350–510 nm. The effect of reaction time and concentration of  $\text{AgNO}_3$  on the synthesis of AgNPs was measured in triplicate.

The morphology of AgNPs was observed using transmission electron microscopy (TEM, FEI, model: TECNAI G2 20). One milliliter of sample solution was diluted 20 times in deionized water, deposited onto 400-mesh carbon-coated Cu grids, and dried at ambient conditions for 24 h. Micrograph analysis was performed at 200 kV acceleration voltage. The surface of the coated paper with AgNPs and high-resolution energy-dispersive X-ray spectroscopy (EDS) mapping were characterized using a scanning electron microscope (SEM, Hitachi Miniscope model TM-3000). All samples were coated with gold using an ion sputtering device.

X-ray diffraction of sample was investigated by using a SmartLab X-ray diffractometer over a diffraction angle range of  $0^{\circ} \leq 2\theta \leq 80^{\circ}$ ; the diffractometer was equipped with a Cu  $K\alpha$  radiation source (wavelength  $\lambda = 1.542 \text{ \AA}$ ) operated on a scan rate of  $10^{\circ} (2\theta)$  at 40 kV and 30 mA.

The structural interaction of samples was analyzed by attenuated total reflection infrared (ATR-FTIR) spectroscopy (Jasco 4200). The spectra of all samples were tested in the range of  $4000\text{--}400 \text{ cm}^{-1}$  with 64 scans at a resolution of  $2 \text{ cm}^{-1}$ .

The antimicrobial activity of the sample was evaluated by agar diffusion. Gram-negative and gram-positive bacteria, *i.e.*, *E. coli* and *S. aureus*, respectively, were selected as representative strains of food pathogens. The bacteria were cultured overnight in brain heart fusion broth at 37  $^{\circ}\text{C}$ . The bacterial culture was diluted in a medium to obtain turbidity of approximately  $10^8 \text{ CFU/mL}$ . The bacterial inoculums were seeded on Muller Hinton Agar plates using the swab plate technique. After 24 h of incubation at 37  $^{\circ}\text{C}$ , the inhibition zone (mm) around the sample disk was measured in triplicate.

Dengue virus serotype 3 (DENV-3) strain P12/08 and Vero cell were used to determine the antiviral activity of the coated paper. The antiviral activity of the sample was investigated using immunofocus assays. Vero cells were seeded in 96-well plates at  $2 \times 10^4$  cells/well in Dulbecco's modified eagle medium (DMEM) supplement with 10% fetal bovine serum (FBS) and incubated at 37  $^{\circ}\text{C}$  in 5%  $\text{CO}_2$  overnight. Samples were cut (1 cm  $\times$  1 cm) and sterilized by pressurized saturated steam at 121  $^{\circ}\text{C}$  for 15 min using Autoclave. The sterile coated paper was placed into the bottom of sterile 1.5-mL microcentrifuge tubes. One hundred microliters of DENV-3 were added to each coated paper sample. After adsorption, 200  $\mu\text{L}$  of DMEM without FBS was immediately added into the coated paper samples and incubated at room temperature for 0, 15, 30, 60, and 120 min. After being incubated at various time points, the coated paper was discarded. The viral treated supernatant was serially diluted in DMEM without FBS from  $10^{-1}$  to  $10^{-4}$  in tenfold increments.

Each dilution was inoculated into Vero cell (100  $\mu\text{L}/\text{well}$ ) and incubated for 2 h at 37  $^{\circ}\text{C}$  in 5%  $\text{CO}_2$ . After 2 h of incubation, the cells were overlaid with Eagle minimal essential medium containing 1.5% methylcellulose (FUJIFILM, Wako Pure Chemical Corporation) and 2% FBS. Three days after infection, the cells were fixed with 3.7% v/v formaldehyde/phosphate-buffered saline (PBS) for 1 h and permeabilized with 0.5% v/v Triton X-100/PBS (Sigma, USA) for 10 min. After 1 h of incubation in a blocking buffer containing 1% FBS and 0.05% Tween-20 in PBS, the cells were treated with a mouse monoclonal antibody 4G2 for 1 h and washed three times with PBS. Then, the cells were incubated with Alexa Fluor 488 goat anti-mouse IgG for 1 h in blocking buffer. Finally, the cells were washed three times with PBS, and the number of foci was counted under a fluorescence microscope (Olympus).

The hydrophobicity of the coated paper with AgNPs was measured by using an optical contact angle meter (OCA 15EC) with 5  $\mu\text{L}$  of deionized water droplet at ambient temperature [32], and an average of five measurements per sample was reported. Water vapor transmission rate (WVTR) of the coated paper was also tested. A sample with 10 mL of distilled water was weighed and kept in a desiccator at 25  $^{\circ}\text{C}$  and 52% RH. The weight gain of sample was measured as a function of time. A slope of linear portion of the weight gained versus time was a value of WVTR. The difference in partial pressure of water vapor across the sample ( $\Delta P$ ) was also determined. The water vapor permeability (WVP) was then calculated following Eq. (1).

$$WVP = \frac{WVTR \times \text{Thickness}}{\Delta P} \quad (1)$$

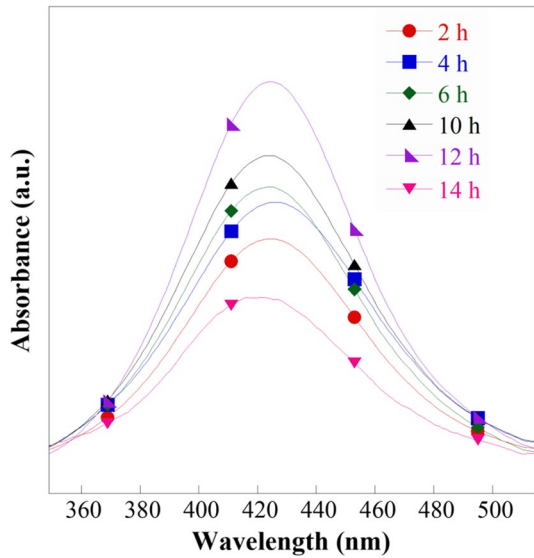
The tensile strength of the coated paper was tested using a universal testing machine (Shimadzu SCG) equipped with a 5 kN load cell following the ASTM D 882-10 method (2015). The initial distance among the grips was 50 mm, and a crosshead speed of 5  $\text{mm}\cdot\text{s}^{-1}$  was used.

## Results and discussion

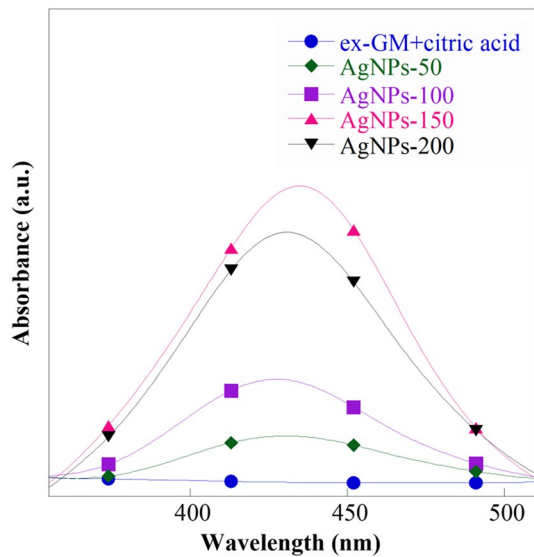
### Synthesis and characterization of AgNPs for paper coating

The formation of AgNPs as a function of reaction time was monitored using UV–vis spectroscopy (Fig. 1). The characteristic surface plasmon resonance (SPR) peaks were observed around 420–424 nm at different reaction times, which indicated the formation of AgNPs [21]. Moreover, the change in peak intensities was observed when the reaction time increased from 2 to 12 h. The increase in peak intensity indicated the increase in the concentration of AgNPs [33]. After 12 h of reaction, no increase in peak intensity was observed. The broadened SPR peak and decrease in magnitude were observed. This phenomenon implied that longer reaction time could lead to the aggregation of AgNPs [34, 35]. Therefore, the reaction was completed within 12 h. This suitable reaction time was selected when synthesizing AgNPs at various silver nitrate concentrations. Figure 2 shows the UV–Vis spectrum of

**Fig. 1** UV–Vis spectrum of green synthesized AgNPs in presence of different reaction time



**Fig. 2** UV–Vis spectrum of green synthesized AgNPs at different concentrations of silver nitrate



green-synthesized AgNPs at 0–200 mM of silver nitrate. The shift of the absorbance peak to a shorter wavelength was observed in AgNPs-100 when compared with AgNPs-50. The blue shift of the wavelength indicated the decrease in particle size [36]. For AgNPs-150 and AgNPs-200, the absorbance peaks shifted to a longer wavelength, which referred to the red shift and implied the increase in particle size [37]. Dutta et al. [38] suggested that the change in reducing environment during the green synthesis of AgNPs resulted in the shift of absorbance peak. However, the absorbance peak of AgNPs-150 showed a red shift, and it still had the highest peak

intensity. A higher peak intensity indicated higher formation of AgNPs [37]. The decrease in peak intensity was found in AgNPs-200. The use of high silver nitrate concentration in green synthesis resulted in a large number of AgNPs formed with a high surface area and surface tension, which led to the agglomeration of AgNPs [39]. The obtained UV–Vis spectrum results were consistent with the TEM analysis (Fig. 3). Particles with an almost spherical shape were also found in all AgNP samples (Fig. 3a). The amount of AgNPs tended to increase with the increase of silver nitrate concentration. The slightly aggregated particles were found in AgNPs-150, whereas AgNPs-200 had densely aggregated particles. Figure 3b exhibits the particle-size distribution of AgNPs. The average particle size could be divided into two ranges. The first range was  $2.36 \pm 0.87$ – $3.74 \pm 1.11$  nm for AgNPs-50 and AgNPs-100, whereas the second range was  $189.96 \pm 71.27$ – $294.73 \pm 106.46$  nm for AgNPs-150 and AgNPs-200.

The crystalline structure of AgNPs on the coated paper was characterized using XRD. The XRD pattern of Ag crystals was observed in the  $2\theta$  range of  $30^\circ$ – $80^\circ$ , which represented indexed reflections of (111), (200), (220), and (311) from the face-centered cubic (FCC) unit cell [40]. Figure 4 shows that  $2\theta$  values of the paper coated with AgNPs were found at  $38.05^\circ$ ,  $47.36^\circ$ , and  $64.77^\circ$  corresponding to the (111), (200), and (220), respectively [41, 42]. Some unknown peaks (marked with a star) might be related to bio-organic phases on the surface of AgNPs [43, 44]. Consequently, the obtained AgNPs on the coated paper had an FCC structure.

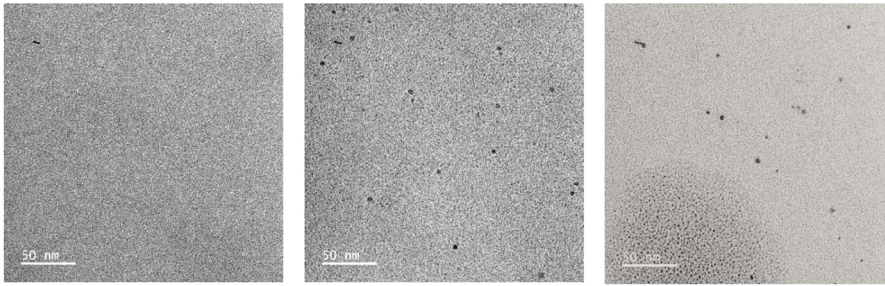
### Attenuated total reflection infrared characterization of paper coated with AgNPs

The ATR-FTIR spectra of papers coated with ex-GM, ex-GM combined with citric acid, and AgNPs-150 are depicted in Fig. 5. The paper coated with ex-GM showed characteristic peaks at  $1013$  and  $1066$   $\text{cm}^{-1}$  (C–O stretching) [45],  $1342$ – $1413$   $\text{cm}^{-1}$  (CH or  $\text{CH}_2$  bending) [46], and  $3263$   $\text{cm}^{-1}$  (OH stretching) [45]. The presence of citric acid in paper coated with ex-GM and AgNPs-150 showed a new peak at  $1706$   $\text{cm}^{-1}$ , which corresponded to carboxyl (C=O) groups and ester linkages of citric acid [22]. The shift of absorbance peaks at  $3263$ – $3306$   $\text{cm}^{-1}$  was also found. This phenomenon was attributed to intermolecular hydrogen bonding among reactants such as PVA, starch, ex-GM, citric acid, and AgNPs. This observation was consistent with the finding of a previous report [45].

### Antibacterial and antiviral activities of paper coated with AgNPs

During the COVID-19 pandemic, various packaging materials have been developed with incorporated antibacterial and antiviral properties. The coating of active compounds on packaging such as film, paper, and cardboard bags could address infection-related concerns of users and promote the safety use of such products [30]. The antibacterial and antiviral activities of paper coated with AgNPs are shown in Figs. 6 and 7, respectively. Figure 6a shows the inhibition zones of *S. aureus* and *E. coli*. The paper coated with AgNPs showed a better antibacterial activity for *E. coli* than for *S. aureus*. Gram-negative bacteria (*E.*

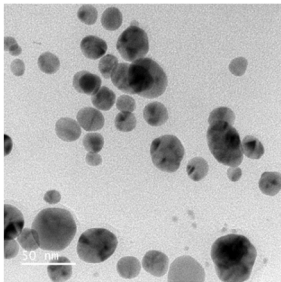
(A)



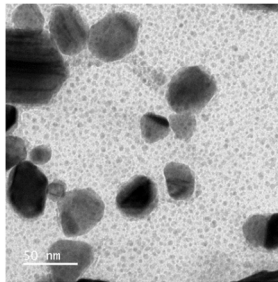
ex-GM+citric acid

AgNPs-50

AgNPs-100

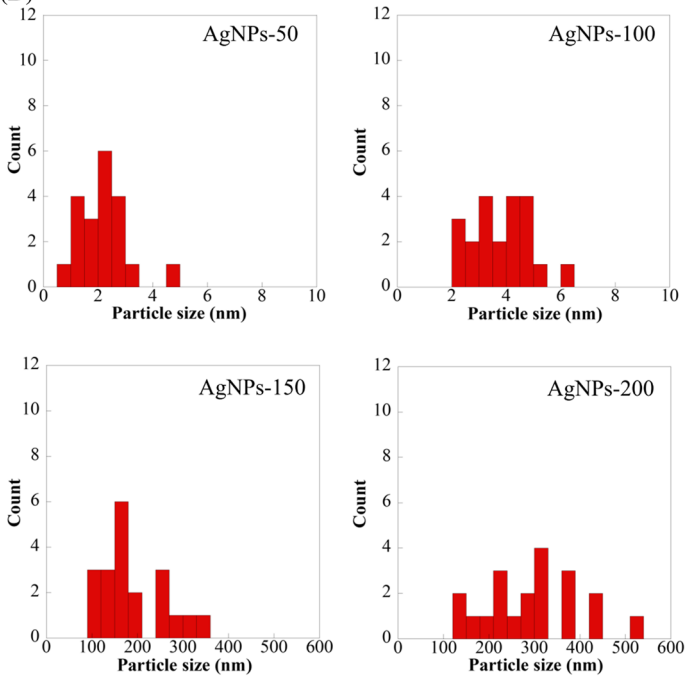


AgNPs-150



AgNPs-200

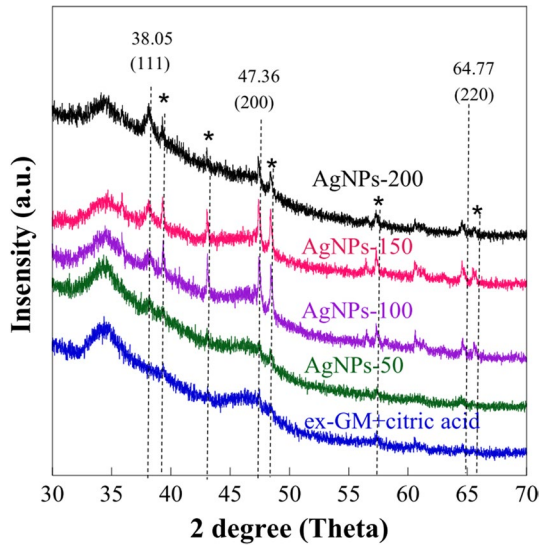
(B)



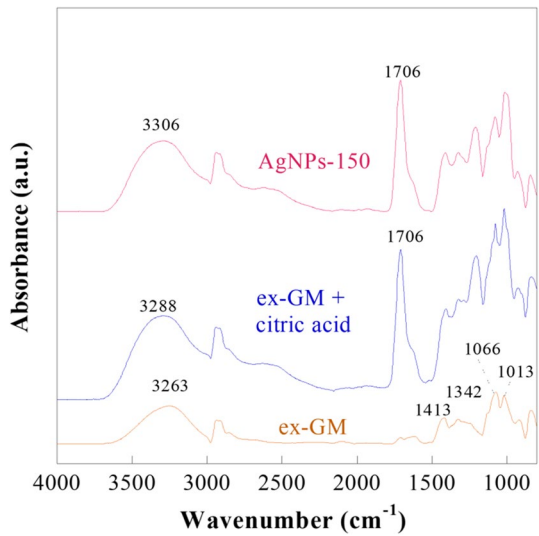
**Fig. 3** A TEM micrographs of AgNPs at different concentrations of silver nitrate and **B** number size distribution of AgNPs in polymer matrix



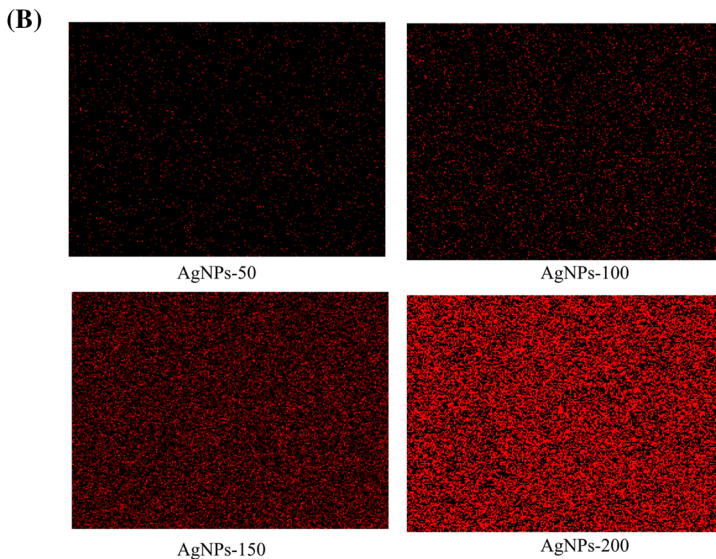
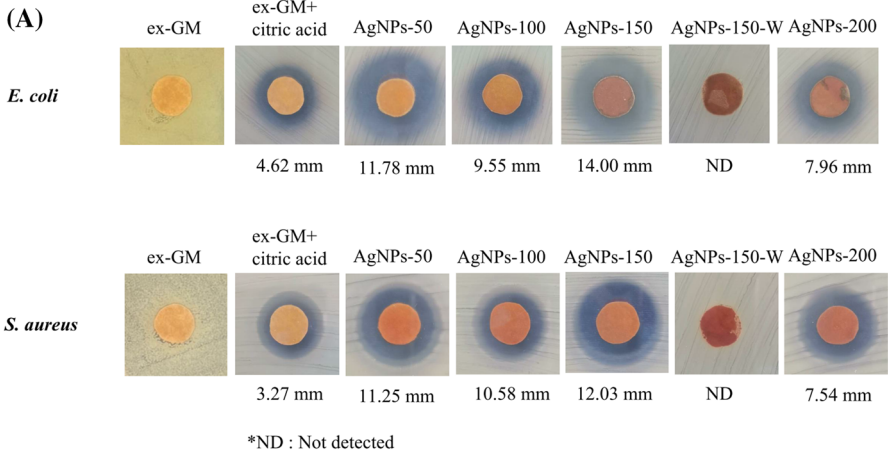
**Fig. 4** XRD pattern of coated paper with AgNPs



**Fig. 5** ATR-FTIR spectra of coated paper with AgNPs



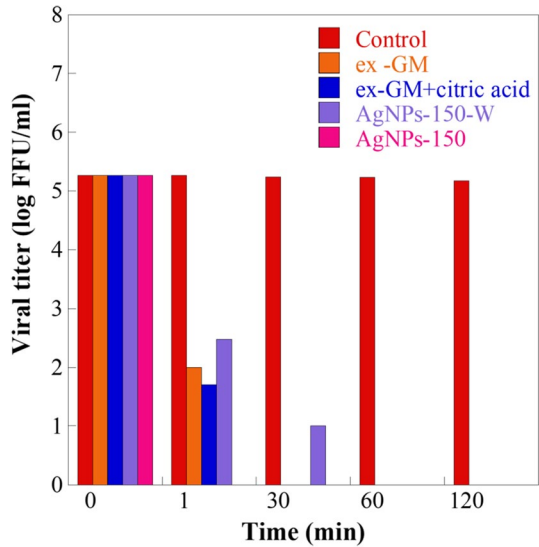
*coli*) had less resistance to inhibition than gram-positive bacteria (*S. aureus*). A thinner peptidoglycan layer of gram-negative bacteria allowed silver ions to penetrate into the cells more easily. Antimicrobial actions of AgNPs have been proposed with four mechanisms, viz. (i) adhesion of AgNPs toward the surface membrane of microbial, (ii) penetration of AgNPs into the cells resulting in the disruption of biomolecules and intracellular damage, (iii) generation of reactive oxygen species for attacking microbial cells, and (iv) disruption of the signal transduction pathways in the cells [47]. Furthermore, other bioactive agents in



**Fig. 6** **A** antibacterial activities of paper coated with AgNPs and **B** EDS images of coated paper with AgNPs

the coating of the paper such as citric acid and ex-GM had antibacterial properties. The main actions of antibacterial activities for citric acid were acidulation and chelation. The acidic ions can damage the enzymes and extracellular membrane and bind essential metal ions in bacterial cells [48]. For ex-GM, the bioactive compounds such as xanthone, saponin, terpenoid, tannin, and flavonoid had an antibacterial activity. Xanthone could retard cell replication; saponin and terpenoid could destroy cell membrane, and tannin could inhibit protein transport enzyme through the cell membrane [49]. In addition, paper coated with AgNPs-150 showed a larger clear zone than paper coated with ex-GM, paper coated with

**Fig. 7** Antiviral activity of paper coated with AgNPs



ex-GM, and citric acid and paper coated with AgNPs-150-W (without citric acid). This observation indicated the synergistic effect of the use of active compound combination, viz. AgNPs, citric acid, and ex-GM, on antibacterial activities. The antibacterial capability of active compounds coated on a paper was in the following increasing order: AgNPs-150-W and ex-GM < citric acid combined with ex-GM < AgNPs-150. In addition, the dispersion ability of AgNPs in a polymer matrix was considered as a factor influencing antibacterial activities. Azeredo [50] proposed that the good dispersion of AgNPs in a polymer matrix increased penetration ability into the cell wall, thereby enhancing the antibacterial activity. The dispersion of AgNPs-150 could be confirmed by EDS images (Fig. 6b). The mapping of silver and element analysis showed a homogeneous distribution of AgNPs-150 on the surface of the paper.

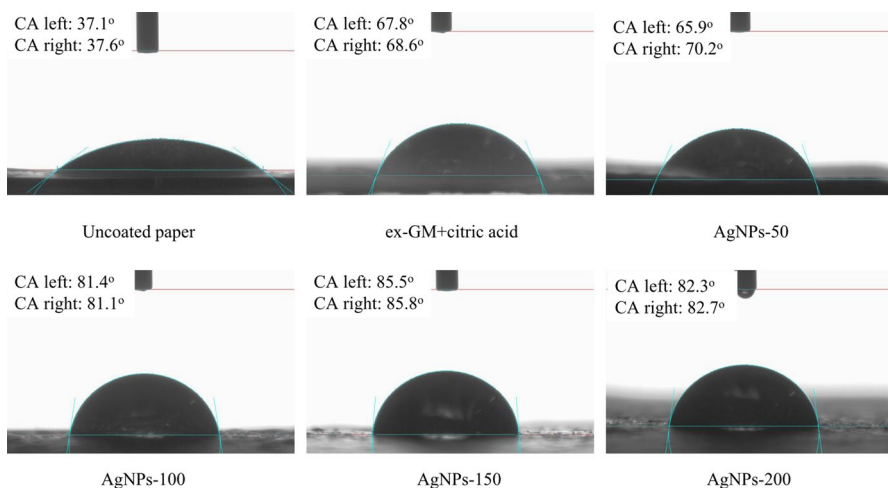
Figure 7 shows the influence of the paper coated with different formulas of AgNPs on antiviral activities. The titer of virus drastically decreased upon contact with paper coated with ex-GM, ex-GM combined with citric acid, AgNPs-150 without citric acid, and AgNPs-150. In addition, the virus titer of the paper coated with AgNPs-150 was found to be zero after 1 min. These results implied that the use of AgNPs combined with ex-GM and citric acid played an important role for effective antiviral properties. For antiviral activities of AgNPs, two mechanisms have been proposed, that is, (i) the binding of AgNPs to the outer surface of virus can inhibit the attachment of virus toward cell receptors, and (ii) AgNPs can bind to the DNA or RNA of the virus resulting in the inhibition of viral replication and propagation inside the host cells [47]. Citric acid can create an acidic environment to inhibit the virus growth [51], whereas the essential compounds in ex-GM could inhibit viral replication [52]. Based on the results of antibacterial and antiviral activities, the paper coated with AgNPs could be applied as packaging for various items such as foods and medical products. However, the

cytotoxicity test of this active packaging should be further studied to confirm and ensure safety.

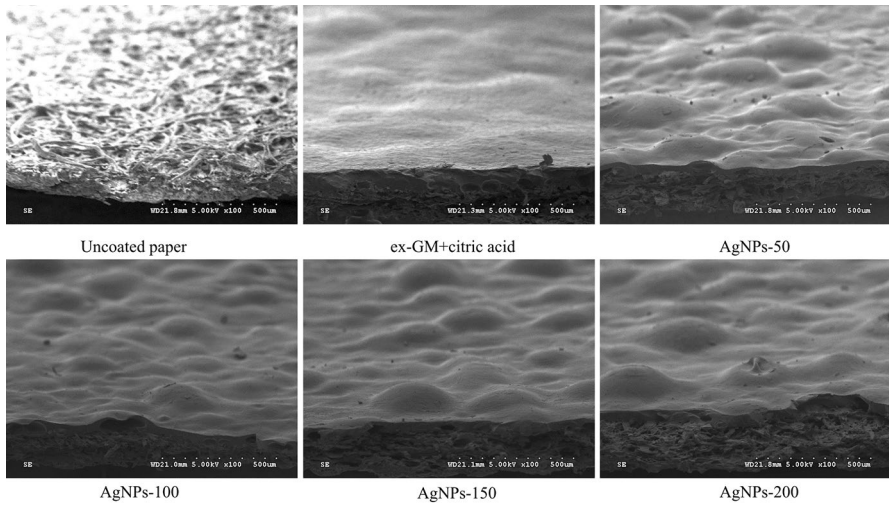
### Physical and mechanical properties of paper coated with AgNPs

However, the poor water resistance of paper limits its applications [53]. The wettability of materials can be investigated by water contact angle (WCA) analysis. The WCA values of paper coated with AgNPs are shown in Fig. 8. The WCA values of paper coated with AgNPs are depicted in Fig. 8. The WCA value remarkably increased from  $\sim 37^\circ$  for uncoated paper to  $81.4^\circ$ – $85.8^\circ$  when paper was coated with AgNPs-100, AgNPs-150, and AgNPs-200. The increase of WCA implied the enhancement of hydrophobicity [54]. Wetting of any surface was based on two main factors, viz. chemical composition on the surface and surface roughness [55]. Figure 9 exhibits SEM images showing the roughness of the samples. The paper coated with AgNPs-100, AgNPs-150, and AgNPs-200 showed a rougher surface than the other samples. Based on Cassie's theory, water droplet holds on the top pyramid shape, whereas the grooves will be occupied with air. This phenomenon can prevent water from filling in the grooves [55, 56]. Therefore, the roughness of the paper coated with AgNPs should be further studied to consolidate architecture and surface morphology by atomic force microscopy.

WVP is a main packaging property that indicates resistance capability to water vapor transmission of packaging materials. A low WVP is required to minimize the moisture transfer from the surrounding to the packaged product, particularly under harsh moisture conditions [57]. The WVP of samples is illustrated in Table 1. The WVP decreased from  $5.84 \pm 0.71 \times 10^{-8} \text{ g/m}^2 \times \text{kPa} \times \text{h}$  for uncoated paper to  $0.17 \pm 0.68 \times 10^{-8}$ – $0.20 \pm 0.31 \times 10^{-8} \text{ g/m}^2 \times \text{kPa} \times \text{h}$  for coated paper. Srichiangsa et al. [58] suggested that the coating of paper could reduce the number and size of



**Fig. 8** WCA values of paper coated with AgNPs



**Fig. 9** SEM images of coated paper with AgNPs

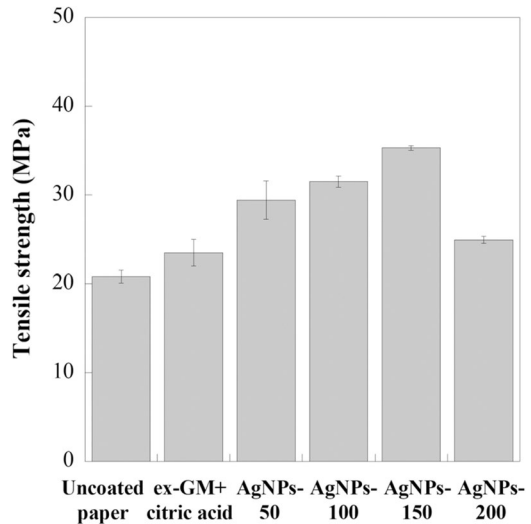
**Table 1** WVP of uncoated and coated paper

Sample	WVP ( $\text{g}/\text{m}^2 \times \text{kPa} \times \text{h}$ )
Uncoated paper	$5.84 \pm 0.71 \times 10^{-8}$
ex-GM combined with citric acid	$0.17 \pm 0.68 \times 10^{-8}$
AgNPs-50	$0.18 \pm 0.19 \times 10^{-8}$
AgNPs-100	$0.19 \pm 2.37 \times 10^{-8}$
AgNPs-150	$0.20 \pm 2.18 \times 10^{-8}$
AgNPs-200	$0.20 \pm 0.31 \times 10^{-8}$

pores on the paper surface, resulting in the poor permeability of the paper package to water vapor. Furthermore, the presence of nanoparticles in coated paper could reduce WVP by the formation of a tortuous path for water vapor transmission.

The tensile strength of paper coated with AgNPs is depicted in Fig. 10. The tensile strength indicates the maximum stress in which the paper packaging can withstand before breaking [55]. All the coated papers showed higher values in tensile strength when compared with uncoated paper. The tensile strength increased up to 69.58% when the paper was coated with AgNPs-150. The formation of strong intermolecular hydrogen bonds between the polymer matrix and AgNPs could enhance tensile strength [59, 60]. The obtained tensile strength at 35.27 MPa for paper coated with AgNPs-150 was comparable to other common packaging paper in previous reports, for example, paper coated with curdlan/chitosan (25–37 MPa) [61], paper coated with chitosan–caseinate bilayer (21–26 MPa) [62], and paper coated with carboxymethyl cellulose/cellulose nanocrystal-immobilized AgNPs (18–20 MPa) [60]. However, increasing

**Fig. 10** Tensile strength of paper coated with AgNPs



AgNP concentration at AgNPs-200 for coating, the tensile strength decreased to 24.93 MPa. Vaezi et al. [63] indicated that the agglomeration of nanoparticles might result in the decrease of tensile strength.

## Conclusion

Bioactive packaging based on a paper coated with green-synthesized AgNPs having antiviral and antibacterial activities has been successfully developed. The AgNPs were prepared from different reaction times and  $\text{AgNO}_3$  concentration using ex-GM as a bio-reducing agent. The optimal reaction time was 12 h. The result showed that the synthesized AgNPs with various  $\text{AgNO}_3$  concentration were mostly spherical in shape with an average particle size of 2.36–294.73 nm. Based on silver mapping and element analysis, AgNPs-150 had a homogeneous distribution on the paper surface. The paper coated with AgNPs showed greater hydrophobicity because of the greater surface roughness of the samples. The highest tensile strength of the coated paper was observed in AgNPs-150. The paper coated with AgNPs-150 exhibited the highest antibacterial property against *E. coli* and *S. aureus*, and it also had antiviral properties. Furthermore, the use of combination active compounds, that is, AgNPs, citric acid, and ex-GM, provided the synergistic effect on antibacterial activities. Based on the obtained results, the coated paper with green-synthesized AgNPs integrated with citric acid and ex-GM is a potential material for bioactive packaging application.

**Acknowledgements** This research project is supported by National Research Council of Thailand (NRCT): NRCT5-RGJ63003-049 and Research and Graduate Studies Khon Kaen University, Thailand.

## References

1. Jung J, Raghavendra GM, Kim D, Seo J (2018) One-step synthesis of starch-silver nanoparticle solution and its application to antibacterial paper coating. *Int J Biol Macromol* 107:2285–2290. <https://doi.org/10.1016/j.ijbiomac.2017.10.108>
2. Tsai TT, Huang TH, Chang CJ et al (2017) Antibacterial cellulose paper made with silver-coated gold nanoparticles. *Sci Rep* 7:1–10. <https://doi.org/10.1038/s41598-017-03357-w>
3. Gatto M, Ochi D, Yoshida CMP, da Silva CF (2019) Study of chitosan with different degrees of acetylation as cardboard paper coating. *Carbohydr Polym* 210:56–63. <https://doi.org/10.1016/j.carbpol.2019.01.053>
4. Kansal D, Hamdani SS, Ping R et al (2020) Food-safe chitosan–zein dual-layer coating for water- and oil-repellent paper substrates. *ACS Sustain Chem Eng* 8:6887–6897. <https://doi.org/10.1021/acssuschemeng.0c02216>
5. Brodnjak U, Tihole K (2020) Chitosan solution containing zein and essential oil as bio based coating on packaging paper. *Coatings* 10:497. <https://doi.org/10.3390/coatings10050497>
6. Šumiga B, Šumiga B, Ravnjak D, BohPodgornik B (2019) Antimicrobial paper coatings containing microencapsulated cymbopogon citratus oil. *Coatings* 9:470. <https://doi.org/10.3390/coatings9080470>
7. Ounkaew A, Kasemsiri P, Srichiangsa N et al (2021) Green synthesis of nanosilver coating on paper for ripening delay of fruits under visible light. *J Environ Chem Eng* 9:105094. <https://doi.org/10.1016/j.jece.2021.105094>
8. Araujo TF, Silva LP (2022) Completely green synthesis of antimicrobial nanocomposites based on hydrogels containing silver nanoparticles for 3d biofabrication of smart scaffolds. *J Polym Environ* 30:2751–2758. <https://doi.org/10.1007/s10924-021-02368-z>
9. Lorwanishpaisarn N, Srikhao N, Jetsrisuparb K et al (2022) Self-healing ability of epoxy vitrimer nanocomposites containing bio-based curing agents and carbon nanotubes for corrosion protection. *J Polym Environ* 30:472–482. <https://doi.org/10.1007/s10924-021-02213-3>
10. Srikhao N, Kasemsiri P, Ounkaew A et al (2021) Bioactive nanocomposite film based on cassava starch/polyvinyl alcohol containing green synthesized silver nanoparticles. *J Polym Environ* 29:672–684. <https://doi.org/10.1007/s10924-020-01909-2>
11. Guerraf A, Jadi S, Bakirhan NK et al (2022) Antibacterial activity and volatile organic compounds sensing property of polypyrrole-coated cellulosic paper for food packaging purpose. *Polym Bull.* <https://doi.org/10.1007/s00289-021-04041-w>
12. Ounkaew A, Kasemsiri P, Jetsrisuparb K et al (2020) Synthesis of nanocomposite hydrogel based carboxymethyl starch/polyvinyl alcohol/nanosilver for biomedical materials. *Carbohydr Polym* 248:116767. <https://doi.org/10.1016/j.carbpol.2020.116767>
13. Vokhidova NR, Rashidova SS (2021) The influence of synthesis conditions on the film morphology of chitosan-stabilized silver nanoparticles. *Polym Bull* 79:3419–3436. <https://doi.org/10.1007/s00289-021-03669-y>
14. Gurme ST, Aware CB, Surwase SN et al (2019) Synthesis of melanin mediated silver nanoparticles from *aeromonas* sp. SNS using response surface methodology: characterization with the biomedical applications and photocatalytic degradation of brilliant green. *J Polym Environ* 27:2428–2438. <https://doi.org/10.1007/s10924-019-01529-5>
15. Dash KK, Kumar A, Kumari S, Malik MA (2021) Silver nanoparticle incorporated flaxseed protein-alginate composite films: effect on physicochemical, mechanical, and thermal properties. *J Polym Environ* 29:3649–3659. <https://doi.org/10.1007/s10924-021-02137-y>
16. GünGök Z, Günay K, Arslan M et al (2020) Coating of modified poly(ethylene terephthalate) fibers with sericin-capped silver nanoparticles for antimicrobial application. *Polym Bull* 77:1649–1665. <https://doi.org/10.1007/s00289-019-02820-0>
17. Priya S, Murali A, Preeth DR et al (2021) Green synthesis of silver nanoparticle-embedded poly(methyl methacrylate-co-methacrylic acid) copolymer for fungal-free leathers. *Polym Bull* 79:4607–4626. <https://doi.org/10.1007/s00289-021-03714-w>
18. Rathore HS, Sivagnanam UT, Abraham LS et al (2021) Green synthesized silver nanoparticles-impregnated novel gum kondagogu–chitosan biosheet for tissue engineering and wound healing applications. *Polym Bull* 79:7215–7227. <https://doi.org/10.1007/s00289-021-03832-5>
19. Hashemabadi M, Sasan H, Amandadi M et al (2021) Natural gum as bio-reductant to green synthesize silver nanoparticles: assessing the apoptotic efficacy on MCF-7 and SH-SY5Y cell



- lines and their antimicrobial potential. *Polym Bull* 78:2867–2886. <https://doi.org/10.1007/s00289-020-03238-9>
20. Jasiūnas L, McKenna ST, Bridžiuvienė D, Miknius L (2020) Mechanical, thermal properties and stability of rigid polyurethane foams produced with crude-glycerol derived biomass biopolyols. *J Polym Environ* 28:1378–1389. <https://doi.org/10.1007/s10924-020-01686-y>
  21. Taesuwan I, Ounkaew A, Okhawilai M et al (2021) Smart conductive nanocomposite hydrogel containing green synthesized nanosilver for use in an eco-friendly strain sensor. *Cellulose* 29:273–286. <https://doi.org/10.1007/s10570-021-04302-x>
  22. Ounkaew A, Janaum N, Kasemsiri P et al (2021) Synergistic effect of starch/polyvinyl alcohol/citric acid films decorated with in-situ green-synthesized nano silver on bioactive packaging films. *J Environ Chem Eng* 9:106793. <https://doi.org/10.1016/j.jece.2021.106793>
  23. Rangam NV, Sudagar AJ, Ruszczak A et al (2022) Valorizing the unexplored filtration waste of brewing industry for green silver nanocomposite synthesis. *Nanomaterials* 12:442. <https://doi.org/10.3390/nano12030442>
  24. Naganathan K, Thirunavukkarasu S (2017) Green way genesis of silver nanoparticles using multiple fruit peels waste and its antimicrobial, anti-oxidant and anti-tumor cell line studies. *IOP Conf Series Mater Sci Eng* 191:012009
  25. Mostafa YS, Alamri SA, Alrumman SA et al (2021) Green synthesis of silver nanoparticles using pomegranate and orange peel extracts and their antifungal activity against *Alternaria solani*, the causal agent of early blight disease of tomato. *Plants* 10:2363. <https://doi.org/10.3390/plants10112363>
  26. Akao Y, Nakagawa Y, Nozawa Y (2008) Anti-cancer effects of xanthenes from pericarps of mango-steen. *Int J Mol Sci* 9:355–370. <https://doi.org/10.3390/ijms9030355>
  27. Lee KX, Shameli K, Mohamad SE et al (2019) Bio-mediated synthesis and characterisation of silver nanocarrier, and its potent anticancer action. *Nanomaterials* 9:1423. <https://doi.org/10.3390/nano9101423>
  28. Owoicho O, Olwal CO, Quaye O (2021) Potential of laser-induced fluorescence-light detection and ranging for future stand-off virus surveillance. *Microb Biotechnol* 14:126–135. <https://doi.org/10.1111/1751-7915.13698>
  29. Mizielińska M, Nawrotek P, Stachurska X et al (2021) Packaging covered with antiviral and antibacterial coatings based on ZnO nanoparticles supplemented with geraniol and carvacrol. *Int J Mol Sci* 22:1717. <https://doi.org/10.3390/ijms22041717>
  30. Ordon M, Nawrotek P, Stachurska X, Mizielińska M (2021) Polyethylene films coated with antibacterial and antiviral layers based on CO<sub>2</sub> extracts of raspberry seeds, of pomegranate seeds and of rosemary. *Coatings* 11:1179. <https://doi.org/10.3390/coatings11101179>
  31. Trongchuen K, Ounkaew A, Kasemsiri P et al (2018) Bioactive starch foam composite enriched with natural antioxidants from spent coffee ground and essential oil. *Starch/Staerke* 70:1–9. <https://doi.org/10.1002/star.201700238>
  32. Lorwanishpaisarn N, Kasemsiri P, Srikhao N et al (2019) Fabrication of durable superhydrophobic epoxy/cashew nut shell liquid based coating containing flower-like zinc oxide for continuous oil/water separation. *Surf Coatings Technol* 366:106–113. <https://doi.org/10.1016/j.surfcoat.2019.03.021>
  33. Cheng K, Hung Y, Chen C et al (2014) Green synthesis of chondroitin sulfate-capped silver nanoparticles: characterization and surface modification. *Carbohydr Polym* 110:195–202. <https://doi.org/10.1016/j.carbpol.2014.03.053>
  34. Ortega-Arroyo L, Martín-Martínez ES, Aguilar-Mendez MA et al (2013) Green synthesis method of silver nanoparticles using starch as capping agent applied the methodology of surface response. *Starch/Staerke* 65:814–821. <https://doi.org/10.1002/star.201200255>
  35. Filippo E, Manno D, Buccolieri A et al (2012) Green synthesis of sucralose-capped silver nanoparticles for fast colorimetric triethylamine detection. *Sens Actuat B Chem* 178:1–9. <https://doi.org/10.1016/j.snb.2012.12.061>
  36. Ajitha B, Kumar Reddy YA, Reddy PS et al (2016) Role of capping agents in controlling silver nanoparticles size, antibacterial activity and potential application as optical hydrogen peroxide sensor. *RSC Adv* 6:36171–36179. <https://doi.org/10.1039/c6ra03766f>
  37. Affes S, Maalej H, Aranaz I et al (2020) Controlled size green synthesis of bioactive silver nanoparticles assisted by chitosan and its derivatives and their application in biofilm preparation. *Carbohydr Polym* 236:116063. <https://doi.org/10.1016/j.carbpol.2020.116063>



38. Dutta T, Ghosh NN, Das M et al (2020) Green synthesis of antibacterial and antifungal silver nanoparticles using *Citrus limetta* peel extract: Experimental and theoretical studies. *J Environ Chem Eng* 8:104019. <https://doi.org/10.1016/j.jece.2020.104019>
39. Rattanaaruengsrikul V, Pimpha N, Supaphol P (2012) In vitro efficacy and toxicology evaluation of silver nanoparticle-loaded gelatin hydrogel pads as antibacterial wound dressings. *J Appl Polym Sci* 124:1668–1682. <https://doi.org/10.1002/app.35195>
40. Kamal T, Khan MSJ, Khan SB et al (2020) Silver nanoparticles embedded in gelatin biopolymer hydrogel as catalyst for reductive degradation of pollutants. *J Polym Environ* 28:399–410. <https://doi.org/10.1007/s10924-019-01615-8>
41. Shankar S, Chorachoo J, Jaiswal L, Voravuthikunchai SP (2014) Effect of reducing agent concentrations and temperature on characteristics and antimicrobial activity of silver nanoparticles. *Mater Lett* 137:160–163. <https://doi.org/10.1016/j.matlet.2014.08.100>
42. Gashti MP, Ghehi ST, Arekhloo SV et al (2015) Electromagnetic shielding response of UV-induced polypyrrole/silver coated wool. *Fibers Polym* 16:585–592. <https://doi.org/10.1007/s12221-015-0585-9>
43. Khan MR, Hoque SM, Hossain KFB et al (2020) Green synthesis of silver nanoparticles using *Ipomoea aquatica* leaf extract and its cytotoxicity and antibacterial activity assay. *Green Chem Lett Rev* 13:303–315
44. Awwad AM, Salem NM, Abdeen AO (2013) Green synthesis of silver nanoparticles using carob leaf extract and its antibacterial activity. *Int J Ind Chem* 4:1–6. <https://doi.org/10.1186/2228-5547-4-29>
45. Gürlür N, Paşa S, Erdoğan Ö, Çevik O (2021) Physicochemical properties for food packaging and toxicity behaviors against healthy cells of environmentally friendly biocompatible starch/citric acid/polyvinyl alcohol biocomposite films. *Starch*. <https://doi.org/10.1002/star.202100074>
46. Singha AS, Kapoor H (2014) Effects of plasticizer/cross-linker on the mechanical and thermal properties of starch/PVA blends. *Iran Polym J* 23:655–662. <https://doi.org/10.1007/s13726-014-0260-9>
47. Salleh A, Naomi R, Utami ND et al (2020) The potential of silver nanoparticles for antiviral and antibacterial applications: a mechanism of action. *Nanomaterials* 10:1566. <https://doi.org/10.3390/nano10081566>
48. Ounkaew A, Kasemsiri P, Kamwilaisak K et al (2018) Polyvinyl alcohol (PVA)/starch bioactive packaging film enriched with antioxidants from spent coffee ground and citric acid. *J Polym Environ* 26:3762–3772. <https://doi.org/10.1007/s10924-018-1254-z>
49. Sitti RHS, Sugita P, Ambarsari L, Rahayu DUC (2018) Antibacterial Mangosteen (*Garcinia mangostana* Linn.) peel extract encapsulated in Chitosan. *J Phys Conf Series* 1116:042037
50. de Azeredo HMC (2009) Nanocomposites for food packaging applications. *Food Res Int* 42:1240–1253. <https://doi.org/10.1016/j.foodres.2009.03.019>
51. Aghebaty-Maleki L, Hasannezhad P, Abbasi A, Khani N (2021) Antibacterial, antiviral, antioxidant, and anticancer activities of postbiotics: a review of mechanisms and therapeutic perspectives. *Biointerface Res Appl Chem* 12:2629–2654. <https://doi.org/10.33263/BRIAC122.26292645>
52. Rizaldy D, Hartati R, Nadhifa T, Fidrianny I (2021) Chemical compounds and pharmacological activities of Mangosteen (*Garcinia mangostana* L.) updated review. *Biointerface Res Appl Chem* 12:2503–2526. <https://doi.org/10.33263/BRIAC122.25032516>
53. Jin K, Tang Y, Liu J et al (2021) Nanofibrillated cellulose as coating agent for food packaging paper. *Int J Biol Macromol* 168:331–338. <https://doi.org/10.1016/j.ijbiomac.2020.12.066>
54. Bakhsheshi HR, Ismail AF, Aziz M et al (2020) Co-incorporation of graphene oxide/silver nanoparticle into poly-L-lactic acid fibrous: a route toward the development of cytocompatible and antibacterial coating layer on magnesium implants. *Mater Sci Eng C* 111:110812. <https://doi.org/10.1016/j.msec.2020.110812>
55. Mishra VK, Saini R, Kumar N (2021) A review on superhydrophobic materials and coating techniques. *IOP Conf Series Mater Sci Eng* 1168:012026
56. Radwan AB, Mohamed AMA, Abdullah AM, Maadeed MA (2016) Corrosion protection of electrospun PVDF–ZnO superhydrophobic coating. *Surf Coat Technol* 289:136–143. <https://doi.org/10.1016/j.surfcoat.2015.12.087>
57. Rhim JW, Lee JH, Hong SI (2007) Increase in water resistance of paperboard by coating with poly(lactide). *Packag Technol Sci* 20:393–402. <https://doi.org/10.1002/pts.767>
58. Srichiangsa N, Ounkaew A, Kasemsiri P et al (2022) Facile fabrication of green synthesized silver-decorated magnetic particles for coating of bioactive packaging. *Cellulose* 29:5853–5868. <https://doi.org/10.1007/s10570-022-04636-0>

59. Jung J, Kasi G, Seo J (2018) Development of functional antimicrobial papers using chitosan/starch-silver nanoparticles. *Int J Biol Macromol* 112:530–536. <https://doi.org/10.1016/j.ijbiomac.2018.01.155>
60. He Y, Li H, Fei X, Peng L (2021) Carboxymethyl cellulose/cellulose nanocrystals immobilized silver nanoparticles as an effective coating to improve barrier and antibacterial properties of paper for food packaging applications. *Carbohydr Polym* 252:117156. <https://doi.org/10.1016/j.carbpol.2020.117156>
61. Brodnjak UV (2017) Experimental investigation of novel curdlan/chitosan coatings on packaging paper. *Prog Org Coat* 112:86–92. <https://doi.org/10.1016/j.porgcoat.2017.06.030>
62. Khwaldia K, Basta AH, Aloui H, Saied H (2014) Chitosan–caseinate bilayer coatings for paper packaging materials. *Carbohydr Polym* 99:508–516. <https://doi.org/10.1016/j.carbpol.2013.08.086>
63. Vaezi K, Asadpour G, Sharifi SH (2019) Effect of coating with novel bio nanocomposites of cationic starch/cellulose nanocrystals on the fundamental properties of the packaging paper. *Polym Test* 80:106080. <https://doi.org/10.1016/j.polymertesting.2019.106080>

**Publisher's Note** Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Springer Nature or its licensor holds exclusive rights to this article under a publishing agreement with the author(s) or other rightsholder(s); author self-archiving of the accepted manuscript version of this article is solely governed by the terms of such publishing agreement and applicable law.

## Authors and Affiliations

**Natwat Srikhao<sup>1</sup> · Artjima Ounkaew<sup>1</sup> · Natnaree Srichiangsa<sup>1</sup> ·  
Supranee Phanthanawiboon<sup>2</sup> · Thidarut Boonmars<sup>3</sup> · Atchara Artchayasawat<sup>3</sup> ·  
Somnuk Theerakulpisut<sup>4</sup> · Manunya Okhawilai<sup>5,6</sup> · Pornnapa Kasemsiri<sup>1</sup>**

<sup>1</sup> Department of Chemical Engineering, Faculty of Engineering, Khon Kaen University, Khon Kaen 40002, Thailand

<sup>2</sup> Department of Microbiology, Faculty of Medicine, Khon Kaen University, Khon Kaen 40002, Thailand

<sup>3</sup> Department of Parasitology, Faculty of Medicine, Khon Kaen University, Khon Kaen 40002, Thailand

<sup>4</sup> Energy Management and Conservation Office, Faculty of Engineering, Khon Kaen University, Khon Kaen 40002, Thailand

<sup>5</sup> Metallurgy and Materials Science Research Institute, Chulalongkorn University, Bangkok 10330, Thailand

<sup>6</sup> Research Unit On Polymeric Materials for Medical Practice Devices, Chulalongkorn University, Bangkok 10330, Thailand