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The Potential of Chitosan-*Polygonum minus* Leaf Mediated Silver-nanoparticles Composite as Green Conductive Biofilm

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ABSTRACT

Silver-nanoparticles (AgNPs) from leaf extract have gained considerable interest from years ago until recently. However, the potential of green-synthesised AgNPs as a conductive filler in polymer biocomposites has not been widely investigated. Herein, a series of biopolymer-silver nanoparticle films were prepared by dispersing the suspension of *Polygonum minus* leaf mediated AgNPs into chitosan (CS) matrix via solution casting. In this work, the physicochemical properties of the composite films were evaluated, and structural property was analysed by Fourier transform infrared (FTIR) spectroscopy. The electrical conductivity and surface morphology were investigated by two-point probe and scanning electron microscopy (SEM), respectively. From the evaluation of moisture uptake, solubility and degradation tests, the rate of moisture uptake reduced as AgNPs concentration increased whereas the solubility and degradation rate increased with increasing addition of AgNPs. The FTIR analysis confirmed that there was no new covalent bond formed and suggested that AgNPs interact non-covalently with amine and hydroxyl groups of chitosan matrix. The conductivity of the CS-AgNPs films increased with one-order magnitude from 10^{-8} to 10^{-7} S/cm compared to pristine CS film. The percolation threshold was achieved at 20 wt% of AgNPs and the highest conductivity was achieved at 30 wt% AgNPs with the conductivity value of 3.20 x 10^{-7} S/cm. SEM micrographs revealed that the composite film with 30 wt% AgNPs exhibited smooth and homogeneous surface which agrees well with the conductivity results. This CS-*Polygonum minus* leaf mediated AgNPs composite film shows potential as an alternative for biodegradable biomedical implants, smart packaging and wearable electronics applications.

Keywords: *Chitosan, Silver-nanoparticles, Polygonum minus leaf, Biofilm, Conductive polymer*

1. INTRODUCTION

Biopolymer-based composites have recently attracted significant attention from many researchers not only due to their sustainable, eco-friendly nature, but also because of the ease of processabillity and competitive market of their end products. When this composite is doped with metal nanoparticles such as silver (Ag), the additional antibacterial property and electrical conductivity extend the potential of the films to biomedical applications such as biosensors, biomedical implants and tissue engineering [1-3], smart packaging [4] and textile electronics [5].

The preparation of biopolymer-silver nanoparticle films can be achieved by two methods [6]. The first is *in-situ*, in which polymeric matrix is used as a reaction medium for the formation of AgNPs and functions as a stabilising agent and second is *ex-situ*, in which polymeric matrix is a dispersion medium for independently pre-synthesised AgNPs . On the other hand, plant extracts including leaves, stems, roots, fruits and seeds have been explored in the synthesis of AgNPs as environmentally and economically friendly resources replacing potentially hazardous chemicals like sodium borohydride ($NaBH_4$) and hydrazine [7]. For the nanocomposite films incorporated green-mediated AgNPs, most of the approaches are by *in-situ*. Sadanand et al. [8] generated AgNPs *in-situ* using *Ocimum sanctum* leaf extract as reducing agent in cellulose matrix and obtained eco-friendly nanocomposite films. Nevertheless, an *ex-situ* approach has also been demonstrated. Recently, Ravindran et al. [9] prepared PVA/PEG blend films incorporated AgNPs *ex-situ* using *Capparis zeylanica*. However, none of the work focuses on the investigation of electrical property which is among the desired properties for biomedical applications etc.

In this work, an *ex-situ* generation of biopolymer/ *Polygonum minus* mediated AgNPs composite films was demonstrated by employing chitosan as matrix or dispersion medium and pre-synthesised AgNPs were dispersed and stabilised in the chitosan matrix. *Polygonum minus* is an aromatic plant, known as 'daun kesum' in Malaysia and widely used in cooking as flavouring agent. *Polygonum minus* contains high flavonoid and phenolic content [10] that may act as reducing, stabilizing, and

capping agents in the conversion of silver ions to silver nanoparticles [11].

Meanwhile, chitosan is the second abundant biopolymers. The properties of chitosan such as biocompatibility, biodegradability, and nontoxicity have been extensively proven by researchers through testing with different chemical solvents. Herein, the physicochemical properties were investigated and characterisations by Fourier-transform infrared spectroscopy (FTIR) was carried out. The electrical conductivity was obtained from a two-point probe method and the surface morphology was studied by scanning electron microscopy (SEM) to investigate the effect of AgNPs concentration on their dispersion in the biocomposite films.

2. MATERIALS AND METHODS

Chitosan (medium molecular weight) and acetic acid glacial were purchased from Sigma-Aldrich. Glycerol was purchased from Merck. *Polygonum minus* mediated AgNPs suspensions were obtained from Haiza's research group with an average particle size of 46.8 nm [12]. All reagents used were of analytical grade and used as received without further purification.

2.1. Preparation of AgNPs suspensions

The AgNPs suspensions were prepared by Salim [12] according to the following method. 25 mL of 0.001 M silver nitrate solution was mixed with 20 ml of 0.02 g/ml *Polygonum minus* leaf extract. Subsequently, dropwise of 1.0 M sodium hydroxide that act as pH regulator was added to the complex solution. Then, the complex solution was continuously stirred by using a magnetic stirrer for 20 minutes and kept for 24 hours at room temperature to ensure that the reduction process reached completion.

2.2. Preparation of CS/AgNPs composite films

Chitosan powder was dissolved in 1% (v/v) aqueous solution of acetic acid glacial to make 1 wt% chitosan solution. The solution mixture was agitated under mechanical stirring until homogeneous at room temperature. Then, different concentrations of AgNPs suspensions (10-50 wt%) were added to the pre-prepared chitosan solution and stirring was continued. After CS and AgNPs became homogeneous, 6 ml of glycerol was added as plasticiser. The mixtures were then poured into glass moulds and dried at room temperature for 48 hours to obtain composite thin films.

2.3. Characterisations and Measurements

2.3.1. Moisture Content

The moisture content of the conductive film was determined by using a moisture analyser (HE53 Mettler Toledo, USA). The film was placed into the chamber and moisture content was automatically calculated and average samples data was collected.

2.3.2. Solubility Test

The film samples were cut into the size of 2 × 2 cm and placed in oven for 24 hours at a temperature of 105 °C to obtain the primary dry weight (M_i). Then, each sample was soaked in a container filled with distilled water for 24 hours at a temperature of 25 °C. After this interval, the sample was removed and placed again in the oven for 24 hours at 105 °C and weighed to obtain the secondary dry weight (M_f). The solubility percentage was then calculated using Equation (1) [13].

$$\text{Solubility (\%)} = \left(\frac{Initial\ \ mass\ (Mi) - Final\ \ mass\ (Mf)}{Initial\ \ mass\ (Mi)}\right) \times 100 \quad (1)$$

2.3.3. Degradation Test

The degradation test was conducted in soil and the weight loss of the film was evaluated over time. The weight loss was determined every seven days for 28 days from starting day and was calculated using Equation (2) [14].

$$\text{Weight Loss (\%)} = \left(\frac{Initial\ \ mass - Final\ \ mass}{Initial\ \ mass}\right) \times 100 \quad (2)$$

2.3.4 Structural Analysis by Attenuated Total Reflectance-Fourier Transform Infrared (ATR-FTIR) Spectroscopy

ATR-FTIR spectroscopic analysis of CS and CS/AgNPs films was performed using FTIR spectrophotometer (Perkin-Elmer Spectrum RX1 Series) in the range of 650 to 4000 cm−1 at a resolution of 4 cm−1. The selected FTIR spectra with transmittance % per wavelength (cm-1) were obtained and critical percentage absorbance peak was calculated using Equation (3).

$$\text{Absorbance peak (\%)} = \frac{Initial\ \ Transmittance\ \ - Final\ \ Transmittance}{Initial\ \ Transmittance} \times 100 \quad (3)$$

2.3.5 Electrical Conductivity Measurement

The electrical resistivity of the films was measured using a two-point probe I-V measurement system at room temperature. The measurement was performed using Keithley Model 4200 Semiconductor Characterization System (Keithley Instruments, Cleveland, OH, USA) with voltage ranging from 0 to 1 V. The conductivity (σ) values were obtained from Equation (4), where L is the distance between the electrodes in cm, A is the cross-sectional area in cm2, and R is the electrical resistance in Ω.

$$\text{Conductivity } (\sigma) = \left(\frac{L}{AR}\right) \quad (4)$$

2.3.6 Morphological Study by Scanning Electron Microscopy (SEM)

The AgNPs filler dispersion and distribution in the CS matrix were analysed using a scanning electron microscope, model JOEL JSM-6460LA (Tokyo, Japan). The conductive films were coated with a conductive layer of palladiums about 20

μm thickness by using a sputter coater, model JEOL JFC1600 (Tokyo, Japan) to prevent electrostatic charge during the characterisation.

3. RESULTS AND DISCUSSION

3.1 Physicochemical Properties of CS/AgNPs Films

Table 1 shows the effect of moisture, solubility, and degradation percentage on CS and CS/AgNPs conductive films with different concentrations of AgNPs. The water content in the films were determined by loss from drying, in which the samples were heated, and the weight loss was obtained due to evaporation of moisture. Based on the results, the moisture percentage reduced with increasing of AgNPs concentration. Salari et al. also observed [15] a decrease in moisture content as nanoparticles were added into chitosan films. The increase of compactness in the chitosan matrix has reduced the penetration of water [16].

Table 1. The effect of AgNPs concentration on moisture, solubility, and degradation percentage of CS/AgNPs films

AgNPs (wt%)	Moisture (%)	Solubility (%)	Degradation (%)
0	17.92	28.13	80.46
10	13.58	30.31	89.32
20	11.90	33.56	91.54
30	11.11	35.65	94.64
40	10.43	38.39	97.31
50	10.04	40.34	99.22

From the solubility percentage, the water solubility of the CS film without AgNPs was 28.13 %, and with an increased concentration of AgNPs filler, the water solubility of samples increased. Aradmehr and Jayanbakht [17] also reported that water solubility of chitosan/lignin/silver nanoparticles composite increased by adding silver nanoparticles and it was due to interactions between silver nanoparticles and water. Another researcher reported that solubility of the chitosan/gelatin/AgNPs films slightly increased with the addition of AgNPs that was attributed to the phytochemicals that are water-soluble such as alkaloids, tannins, polyphenols and carbohydrates present on the surface of nanoparticles [18].

After 28 days of exposure in soil, the CS/AgNPs films shrank in size and the films degradation was followed by reduction of overall weight loss within various time intervals. The degradation rate increased with the increasing of AgNPs content. This indicates that moisture from the soil has infiltrated the hydrophilic polymer matrix and degraded the polymer chains and made them easily destroyed by the microorganisms as reported by Ediyilyam et al. [19] who studied the biodegradability of lyophilised chitosan scaffolds produced through a green synthesis method. As the concentration of AgNPs increased, less volume of matrix interacting in soil resulting in higher degradation rate of the conductive films. Both matrix and filler formed a less volume structure, resulting in an easily accessible site for the enzymatic attack [20]. Therefore, it was much easier to undergo the degradation of polymer chains by microorganisms.

3.2 Fourier-Transform Infrared (FTIR) Analysis

The molecular interactions between chitosan matrix and the extracted AgNPs were determined by FTIR spectral analysis. Figure 1 shows the FTIR spectra of pure chitosan film and CS/AgNP biocomposite films containing 10, 30 and 50 wt% of AgNPs. The spectrum of pure chitosan displayed a broad band at 3374 cm^{-1} which can be assigned to stretching vibrations of O-H and N-H groups. The bands at 2852 cm^{-1} and 2917 cm^{-1} are ascribed to asymmetric stretching vibrations of aliphatic C-H bonds in -CH and -CH_2, respectively. The characteristic band of the carbonyl group (C=O) of secondary amide -CONHR appeared at 1730 cm^{-1} and the characteristic bending vibrations of amine group (NH_2) can be discovered at 1592 cm^{-1}. The band at 1414 cm^{-1} corresponds to the symmetric deformation of C-H bond in -CH_3 while the band at 1323 cm^{-1} is assigned to the vibration modes of amide III. The asymmetric vibration of C-O-C bridge can be assigned to 1110 cm^{-1}. The bands near 1055-1037 cm^{-1} are attributed to C=O vibration of the ring COH and CH_2OH. Similar FTIR bands of extracted chitosan were observed by many authors [21-23].

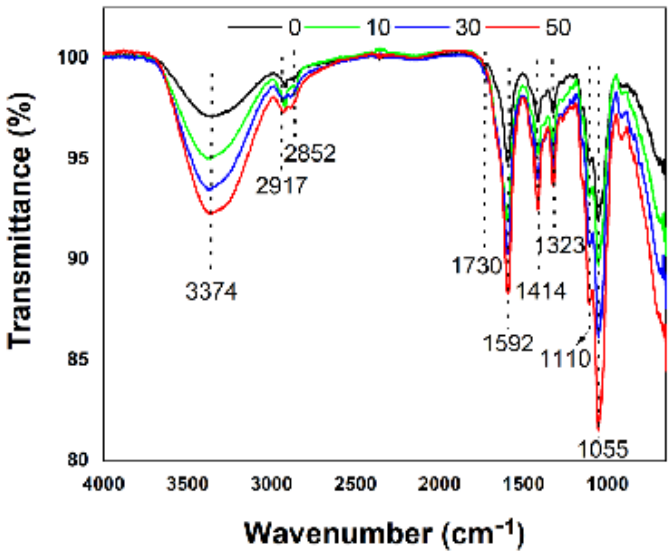

Figure 1. FTIR spectra of pure CS and CS/AgNPs conductive films at 10,30 and 50 wt% of AgNPs.

Figure 1 clearly shows that all these bands broadened, increased in intensity and shifted to longer wavelength in the presence of AgNPs. At 30 and 50 wt% of AgNPs, the C=O band at 1730 cm^{-1} disappeared and the amine band at 1592 cm^{-1} broadened, increased in intensity, and slightly shifted to longer wavelength. Similar observation was also reported by Kaur et al. [24]. This is evidence of Ag^+ complexation with the amine group. Based on the overlapping bands of O-H and N-H at 3364 cm^{-1} the peak intensity increased by 0.22, 0.50, and 0.71 % in the presence of 10, 30 and 50 wt% AgNPs, respectively. The changes were getting prominent as the concentration of AgNPs increased. This

suggests that the silver ions interact with the amine and hydroxyl groups as reported by Gonzalez-Campos et al. [25]. The illustration of van der Waals interactions of silver ions with amine and hydroxyl groups of chitosan is proposed in Figure 2 which is similar to the one proposed by Praveena and Kumar [26] and Nate et al. [27].

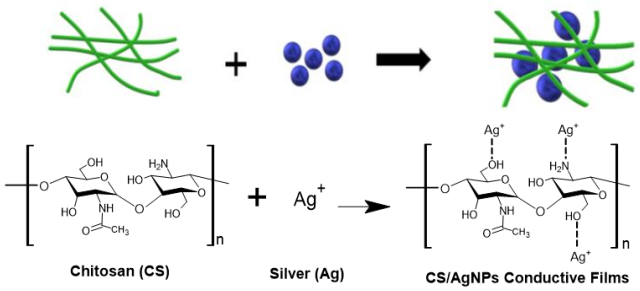

Figure 2. The interactions between CS and Ag ion in CS/AgNPs conductive film.

3.1. Electrical Conductivity

The electrical conductivity of the CS films at different concentration of AgNPs calculated from the obtained resistivity is presented in Figure 3. CS film without AgNPs exhibited low conductivity of 2.30 x 10^{-8} S/cm. From the graph, it is clearly seen that the conductivity began to increase with 10 wt% addition of AgNPs and achieved the percolation threshold at 20 wt% with one-order magnitude of increment. The value at the percolation threshold was 2.03 x 10^{-7} S/cm. The conductivity value continued to increase up to 30 wt% addition of AgNPs with conductivity value of 3.27 x 10^{-7} S/cm. However, the conductivity began to reduce at 40 wt% of AgNPs and reached 2.71 x 10^{-7} S/cm at 50 wt% of the nanofiller. Almost similar trend of conductivity was reported by Hanif et al. [28] when graphite was incorporated into cellulose films. At lower AgNPs fraction, the amount of AgNPs was still insufficient to form conductive percolation network resulting in low conductivity. The interconnected network was gradually formed as the concentration of AgNPs reached 20 wt% due to the optimum formation of charge transfer complexes (CTCs) inside the polymer chain network [29]. Furthermore, further addition of AgNPs up to 40 and 50 wt% has caused agglomeration of AgNPs within the chitosan matrix which has led to a reduction of the values of electrical conductivity [30].

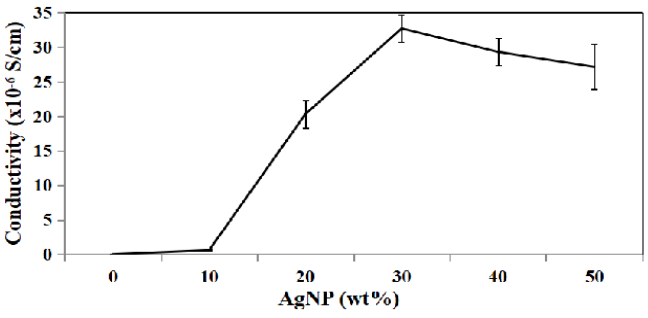

Figure 3. Electrical conductivity of CS/AgNPs films with increasing AgNPs concentration.

3.2. Surface Morphology

Figure 4 represents SEM micrographs of CS/AgNPs conductive films with increasing concentration of AgNPs filler under 200x, 500x, and 1000x magnifications. The micrograph of pure CS demonstrated relatively smooth, compact without any cracks or pores and homogeneous surface although slight excess of chitosan can be observed [31]. The films with the presence of AgNPs also demonstrated smooth surface without macroscopic phase separation indicating that AgNPs were well dispersed in CS matrix. At 10 wt% of AgNPs, the film displayed smoother surface as compared to pure CS. As the concentration of AgNPs increased to 20 wt%, the appearance of white spots was seen clearer, and the dispersion was slightly inhomogeneous. The surface of the film became homogeneous at 30 wt% AgNPs and slight agglomeration was observed for the film with 50 wt% AgNPs. The tendency of agglomerations with increasing AgNPs concentration was also observed by previous reports [32, 33]. The SEM micrographs also suggested that smooth surface of films promote immobilization of Ag+ inside the CS matrix as the highest conductivity was achieved at 30 wt% AgNPs. The surface at 50 wt% AgNPs film was seen rougher and the white spots were more clearly visible. Very likely, the shape of the AgNPs was spherical and it showed tendency to agglomerate into larger cubic and irregular shapes [34].

4. CONCLUSION

Chitosan/silver nanoparticles (CS/AgNPs) conductive biocomposite films with varying concentrations of AgNPs in the range of 10-50 wt% were successfully prepared by the *ex-situ* method via solution casting. The AgNPs were obtained from *Polygonum minus* leaf extract through an eco-friendly approach without the use of hazardous chemicals in the synthesis. The incorporation of AgNPs into the chitosan film decreased the moisture uptake and increased the solubility in water and degradation percentage. Further reduction of moisture uptake and increment of solubility and degradation rate were observed as the concentration of AgNPs increased. The FTIR analysis revealed that weak

interactions occurred between the Ag ion and amine and hydroxyl groups of chitosan matrix which agrees well with the previously reported work [18-20]. The absorbance peaks of amine and hydroxyl group increased with increasing AgNPs filler. The electrical conductivity of the CS/AgNPs films increased one-order magnitude with the addition of 10 wt% AgNPs and the percolation threshold was achieved at 20 wt%. The conductivity value reached its maximum at 30 wt% and was reduced with further addition of AgNPs. The higher increment of AgNPs at 40 and 50 wt% induced the agglomeration on the surface of CS/AgNPs films as displayed by the scanning electron microscope (SEM) micrographs. The findings concluded that the concentration of AgNPs is an important parameter as it influenced the dispersion morphology which relates to electrical conductivity. This study could be useful in the development of polymer-nanocomposites for various applications.

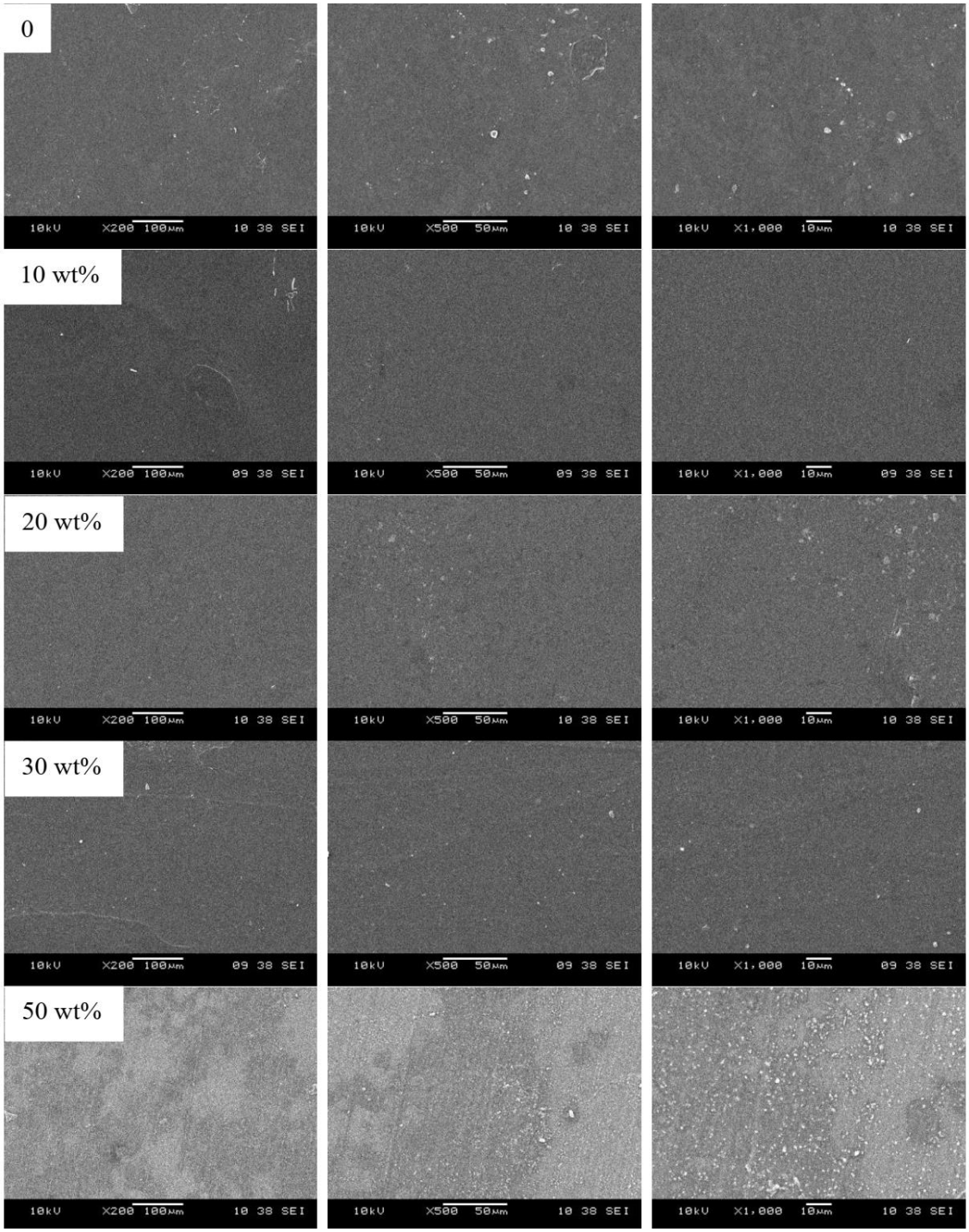

Figure 4. SEM micrographs of CS/AgNPs films with increasing AgNPs concentration.

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REFERENCES

[1] Gradess, R., Abargues, R., Habbou, A., Canet-Ferrer, J., Pedrueza, E., Russell, A., Valdes, J.L. & Martínez-Pastor, J. P. (2009). Localized surface plasmon resonance sensor based on Ag-PVA nanocomposite thin films. Journal of Materials Chemistry, 19(48), 9233-9240.

[2] Divakar, D. D., Jastaniyah, N. T., Altamimi, H. G., Alnakhli, Y. O., Alkheraif, A. A., & Haleem, S. (2018). Enhanced antimicrobial activity of naturally derived bioactive molecule chitosan conjugated silver nanoparticle against dental implant pathogens. International journal of biological macromolecules, 108, 790-797.

[3] Sivashankari, P. R., & Prabaharan, M. (2016). Prospects of chitosan-based scaffolds for growth factor release in tissue engineering. International journal of biological macromolecules, 93, 1382-1389.

[4] Herrera, A., De Avilamontiel, G., & Polocorrales, L. (2018). Chitosan-based films with silver nanoparticles incorporated for food packaging applications. Indian J Sci Technol, 11, 1-6.

[5] Naysmith, A., Mian, N. S., & Rana, S. (2022). Green Synthesised Silver Nanocomposite for Thermoregulating E-Textiles. Engineering Proceedings, 15(1), 15.

[6] Das, R. K., & Das, M. (2019). Study of silver nanoparticle/polyvinyl alcohol nanocomposite. International Journal of Plastics Technology, 23(1), 101-109.

[7] Hebbalalu, D., Lalley, J., Nadagouda, M. N., & Varma, R. S. (2013). Greener techniques for the synthesis of silver nanoparticles using plant extracts, enzymes, bacteria, biodegradable polymers, and microwaves. ACS Sustainable Chemistry & Engineering, 1(7), 703-712

[8] Sadanand, V., Rajini, N., Satyanarayana, B., & Rajulu, A. V. (2016). Preparation and properties of cellulose/silver nanoparticle composites with in situ-generated silver nanoparticles using Ocimum sanctum leaf extract. International Journal of Polymer Analysis and Characterization, 21(5), 408-416.

[9] Ravindran, R. E., Subha, V., & Ilangovan, R. (2020). Silver Nanoparticles blended PEG/PVA Nanocomposites synthesis and characterization for Food packaging. Arabian Journal of Chemistry.

[10] Sumazian, Y., Syahida, A., Hakiman, M., & Maziah, M. (2010). Antioxidant activities, flavonoids, ascorbic acid and phenolic contents of Malaysian vegetables. Journal of Medicinal Plants Research, 4(10), 881-890.

[11] Rajan, R., Chandran, K., Harper, S. L., Yun, S. I., & Kalaichelvan, P. T. (2015). Plant extract synthesized silver nanoparticles: an ongoing source of novel biocompatible materials. Industrial Crops and Products, 70, 356-373.

[12] Salim, N. H. (2021). Green Synthesis of Silver Nanoparticles using Polygonum Minus [Final Year Project Thesis, Universiti Malaysia Perlis].

[13] Barra, A., Ferreira, N. M., Martins, M. A., Lazar, O., Pantazi, A., Jderu, A. A., Neumayer, S. M., Rodriguez, B. J., Enachescu, M., & Ferreira, P. (2019). Eco-friendly preparation of electrically conductive chitosan-reduced graphene oxide flexible bionanocomposites for food packaging and biological applications. Composites Science and Technology, 173, 53–60.

[14] AbdulRasheed-Adeleke, T., Egwim, E. C., Ochigbo, S. S., & Ossai, P. C. (2015). Effect of acetic anhydride and citric acid modification on biodegradability of cassava starch nanocomposite films.

[15] Salari, M., Khiabani, M. S., Mokarram, R. R., Ghanbarzadeh, B., & Kafil, H. S. (2018). Development and evaluation of chitosan based active nanocomposite films containing bacterial cellulose nanocrystals and silver nanoparticles. Food hydrocolloids, 84, 414-423.

[16] Ojagh, S. M., Rezaei, M., Razavi, S. H., & Hosseini, S. M. H. (2010). Development and evaluation of a novel biodegradable film made from chitosan and cinnamon essential oil with low affinity toward water. Food Chemistry, 122(1), 161-166.

[17] Aradmehr, A., & Javanbakht, V. (2020). A novel biofilm based on lignocellulosic compounds and chitosan modified with silver nanoparticles with multifunctional properties: Synthesis and characterization. Colloids and Surfaces A: Physicochemical and Engineering Aspects, 600, 124952.

[18] Ediyilyam, S., George, B., Shankar, S. S., Dennis, T. T., Wacławek, S., Cernik, M., & Padil, V. V. (2021). Chitosan/gelatin/silver nanoparticles composites films for biodegradable food packaging applications. Polymers, 13(11), 1680.

[19] Ediyilyam, S., Lalitha, M. M., George, B., Shankar, S. S., Wacławek, S., Cerník, M., & Padil, V. V. T. (2022). Synthesis, characterization and physicochemical properties of biogenic silver nanoparticle-encapsulated chitosan bionanocomposites. Polymers, 14(3), 463.

[20] Pantani, R., & Sorrentino, A. (2013). Influence of crystallinity on the biodegradation rate of injection-moulded poly (lactic acid) samples in controlled composting conditions. Polymer Degradation and Stability, 98(5), 1089-1096.

[21] El-araby, A., El Ghadraoui, L., & Errachidi, F. (2022). Usage of biological chitosan against the contamination of post-harvest treatment of strawberries by Aspergillus niger. Frontiers in Sustainable Food Systems, 6, 881434.

[22] De Queiroz Antonino, R. S. C. M., Lia Fook, B. R. P., De Oliveira Lima, V. A., De Farias Rached, R. Í., Lima, E. P. N., Da Silva Lima, R. J., et al. (2017). Preparation and characterization of chitosan obtained from Shells of Shrimp (Litopenaeus vannamei Boone).

[23] Kumari, S., Kumar Annamareddy, S. H., Abanti, S., and Kumar Rath, P. (2017). Physicochemical properties and characterization of chitosan synthesized from fish scales, crab and shrimp shells. Int. J. Biol. Macromol. 104, 1697–1705.

[24] Kaur, P., Choudhary, A., & Thakur, R. (2013). Synthesis of chitosan-silver nanocomposites and their antibacterial activity. Int J Sci Eng Res, 4(4), 869.

[25] Gonzalez-Campos, J. B., Mota-Morales, J. D., Kumar, S., Zarate-Trivino, D., Hernandez-Iturriaga, M., Prokhorov, Y., Lepe, M.V., Garcia-Carvajal, Z.Y., Sanchez, I.C. & Luna-Barcenas, G. (2013). New insights into the bactericidal activity of chitosan-Ag bionanocomposite: the role of the electrical conductivity. Colloids and Surfaces B: Biointerfaces, 111, 741-746.

[26] Praveena, V. D., & Kumar, K. V. (2014). Green synthesis of silver nanoparticles from Achyranthes aspera plant extract in chitosan matrix and evaluation of their antimicrobial activities. Indian Journal of Advances in Chemical Science, 2(3), 171-177

[27] Nate, Z., Moloto, M. J., Mubiayi, P. K., & Sibiya, P. N. (2018). Green synthesis of chitosan capped silver nanoparticles and their antimicrobial activity. MRS Advances, 3, 2505-2517.

[28] Hanif, M. P. M., Jalilah, A. J., Badrul, F., & Nuraqmar, S. M. S. (2019, December). The influence of graphite on conductivity, crystallinity and tensile properties of hydroxyethyl cellulose (hec)/graphite composite films. In IOP Conference Series: Materials Science and Engineering (Vol. 701, No. 1, p. 012015). IOP Publishing.

[29] Mahendia, S., Tomar, A. K., & Kumar, S. (2010). Electrical conductivity and dielectric spectroscopic studies of PVA–Ag nanocomposite films. Journal of Alloys and Compounds, 508(2), 406-411.

[30] Mahendia, S., Tomar, A. K., Chahal, R. P., Goyal, P., & Kumar, S. (2011). Optical and structural properties of poly (vinyl alcohol) films embedded with citrate-stabilized gold nanoparticles. Journal of Physics D: Applied Physics, 44(20), 205105.

[31] Hanif, M. P. M., Jalilah, A. J., Anisah, M. F. H., & Tilagavathy, A. (2020). Chitosan/Polyethylene Oxide (PEO) Filled Carbonized Wood Fiber Conductive Composite Film. In Materials Science Forum (Vol. 1010, pp. 638-644). Trans Tech Publications Ltd.

[32] Ediyilyam, S., George, B., Shankar, S. S., Dennis, T. T., Wacławek, S., Černík, M., & Padil, V. V. (2021). Chitosan/gelatin/silver nanoparticles composites films for biodegradable food packaging applications. Polymers, 13(11), 1680.

[33] Kumar, S.; Shukla, A.; Baul, P.P.; Mitra, A.; Halder, D. Biodegradable hybrid nanocomposites of chitosan/gelatin and silver nanoparticles for active food packaging applications. Food Packag. Shelf Life 2018, 16, 178–184.

[34] Tsuji, T., Kikuchi, M., Kagawa, T., Adachi, H., & Tsuji, M. (2017). Morphological changes from spherical silver nanoparticles to cubes after laser irradiation in acetone–water solutions via spontaneous atom transportation process. Colloids and Surfaces A: Physicochemical and Engineering Aspects, 529, 33-37.