Turkish Journal of Agriculture - Food Science and Technology

Synthesis of Silver Nanoparticles using Cellulose and Starch Extracted from Brewer Spent Grain: Assessment of their Antimicrobial and Preservatives Activities

Clement Olusola Ogidi[1,a,*], Ogo Philip Emmanuel[2,b], Olanrewaju Oludotun Daramola[3,c], Mutiat Oyedolapo Bamigboye[4,d], Olu Malomo[5,e]

[1]*Department of Food Science and Technology, School of Agriculture, Food and Natural Resources, Olusegun Agagu University of Science and Technology, Okitipupa, Nigeria*
[2]*Microbiology Unit, Department of Biological Sciences, Kings University, PMB 555, Odeomu, Nigeria*
[3]*Department of Mathematical Science, Kings University, PMB 555, Odeomu, Nigeria*
[4]*Chemistry Unit, Department of Chemical Sciences, Kings University, PMB 555, Odeomu, Nigeria*
[5]*Biotechnology Unit, Department of Biological Sciences, Kings University, PMB 555, Odeomu, Nigeria*
[*]*Corresponding author*

A R T I C L E I N F O	A B S T R A C T
	Non-porous materials like cellulose and starch can be extracted from agro- industrial wastes and incorporated with nanoparticles for effective biotechnological purposes. In this study, silver nanoparticles (AgNps), silver-cellulose nanoparticles (AgNps-C) and silver-starch nanoparticles (AgNps-S) were characterized by UV-visible spectroscopy. Fourier transform infrared spectroscopy (FTIR) was used to identify viable biomolecules involved in capping and active stabilization of AgNps. Average sizes and morphologies of AgNps, AgNps-C and AgNps-S were further analyzed by scanning electron microscopy (SEM) and the percentage composition of each element was investigated by energy-dispersive X-ray spectroscopy (EDS). Antimicrobial activity of the synthesized AgNPs-C and AgNPs-S was tested against multiple antibiotic resistance microorganisms isolated from fish and meat. Zones of inhibition displayed by AgNPs-C and AgNPs-S ranged from 8.00 to 13.30 mm and 5.00 to 10.30 mm, respectively. The Minimum inhibitory concentration (MIC) for AgNPs-C and AgNPS-R ranged from 125 µg/mL to 500 µg/mL and 500 µg/mL to 1000 µg/mL, respectively. AgNPS-S and AgNps-C inhibited the growth of microorganisms associated with spoilage of fish and meat. The bio-applications of AgNP –C and AgNP-S can be exploited in food industries as preservative agent or incorporated to packaging materials to elongate the shelf life of food products and reduce the side effects attributed to chemical preservative agents.

[a] *clementogidi@yahoo.com* http://orcid.org/0000-0002-4154-6750   [b] *emmanuelphilipogo27@gmail.com* https://orcid.org/0000-0001-8381-7427
[c] *oo.daramola@kingsuniversity.edu.ng* https://orcid.org/0000-0001-5548-4434   [d] *oyedolapobamigboye@gmail.com* https://orcid.org/0000-0002-9976-3125
[e] *olumalomo1967@gmail.com* https://orcid.org/0000-0002-4986-0306

Introduction

Agricultural residues such as orange mesocarp, maize stem and cobs, banana and plantain pseudo stem, coconut shell, groundnut husk, sugar cane bagasse, soybean, wood dusts, hull of oath, wheat, rice and brewers' spent grains (BSG) have not been adequately converted to significant purposes and thus, serve as source of pollution to the environment (Israel et al., 2008 and Araújo et al., 2019). On this note, there is a need to focus on importance and bioconversion of agricultural waste biomass into useful biomaterials since they are costless, environmentally friendly and renewable, which will therefore, change the public perception about agro-industrial wastes towards environmental sustainability. Agricultural wastes are ideal renewable resources for production of nanostructures as a substitute to non-biodegradable, carcinogenic and toxic materials (Maraveas, 2020). BSG is a major by-product of

beer industry, representing around 85% of the entire production (Thiago, et al., 2014). BSG contains cellulose; a linear polymer consisting of 6-member ether rings (D-glucose or dextrose) covalently linked together by ether groups called glycosidic bonds and is biodegradable substance that can be extracted for different purposes. Cellulosic and lignocellulosic biomass from agro-wastes are inexpensive, green, differentiated resource and polite for synthesis of valuable nanoporous materials, polymer nanocomposite and nanoparticles (Saini et al., 2015).

Silver nanoparticles (AgNPs) and polymer nanocomposite materials are useful in different areas like medicine, environmental science, pharmaceutical, food, and agricultural industries and in bioremediation (Duncan, 2011; Preetha, et al., 2013 Fariq et al., 2017). Nanoparticle have been noticed antiquity and is currently used in the

control of bacterial and fungal growth to preserve shelf life of foods. AgNPs can be incorporated to biodegradable polymers like cellulose, chitosan, starch, lignin and xylan from different agro-industrial residues to have different type of polymer nano-composites with bio-functionalities and performance properties like mechanical, barrier, optical, thermal, biodegradation, antimicrobial, chemical stability and other functional properties as compared to nanoparticle with no support (Motaung and Linganiso, 2018). Biopolymers derived from lignocellulosic materials always have higher specific strength, modulus, high-volume applications, low cost, low density, low energy consumption, easy processability, renewable origin and possibility of recycling than other composites (Nevárez et al., 2011). Nanomaterials have successfully improved food safety, enhance shelf-life, food packaging and nutritional value as additives without change in taste, food quality and no side effects (He and Hwang, 2016, Das et al., 2017).

Muscle foods such as meat, fish and their products prone to microbial spoilage and serve as primary source of food borne diseases (Lorenzo et al., 2017). Muscle foods contain growth factors that support microbial growth. Bodunde et al. (2019) revealed different microorganisms that associated with spoilage of muscle foods (meat and fish). The action of microbial growth on meat and fish caused spoilage with biochemical and enzymatic deteriorations. The increasing awareness of fish and meat (food) spoilage microorganisms causing diseases with multiple antibiotic resistance is demanding a search for new and efficient antimicrobial agents. Hence, the continuous exploration for effective antimicrobial and preservative agents in food industries has resulted in the emergence and application of bio-nanotechnology (Zorraquín-Peña et al., 2020). Bio-nanotechnology is now an evolving option that can be employed to enhance food bio-functionality, maintain food nutrients and prevent the growth of microorganisms associated with food borne diseases (Srividya et al., 2017 and Bajpai et al., 2018). Degradable bio-polymers combined with nanoparticles could serve as prospective substitutes to conventional antibiotics because of their antimicrobial activities, since they are readily available, simple to prepare, safe, cost effective and eco-friendly. Wastes from cereal grains namely; wheat, rice, corn, oats, barley as well as waste from tubers such as potatoes, yam and cassava establish a very good reducing agent for silver nitrate to silver nanoparticle (Yakout and Mostafa, 2015). Starch have huge possibilities to substitute conventional synthetic products since their modification with other polymers is an attractive source of raw material for production of high-value bio-based products (Temesgen et al., 2021). New biodegradable polymers like chitosan, cellulose and its derivatives, starch, lipids, proteins, polylactic acid or polyvinyl alcohol from renewable biomass, as a single component or in combination with nanomaterial has yielded excellent support to obtain antimicrobial packaging materials (Motelica et al., 2020). This study therefore, assess antimicrobial activity of synthesized silver nanoparticles with cellulose and starch from BSG against microorganisms isolated from fish and meats. The preservatives quality of AgNPs-C and AgNPs-S on meat and fish was also investigated.

Materials and methods

Chemical used
Silver nitrate ($AgNO_3$), Sodium hydroxide (NaOH), sodium chlorite ($NaClO_2$).

Extraction of cellulose
Brewers' spent grain was collected from International Breweries Plc, Ilesa, Nigeria. BSG was washed with distilled water to remove impurities. They were oven dried at 80 °C. It was grounded and sieved, was boiled in 10% NaOH in w/v ratio 1: 20 in an autoclave, the black slurry obtained was filtered and washed using distilled water. The cellulose was filtered out and bleached with 5 % sodium chlorite (NaOCl) for 3 h at 30°C (Pornchai, 2009).

Extraction of starch
BSG was washed with tap water and then with distilled water to remove impurities, water was poured on the BSG and left for 1 hour. The solution was filtered, filtrate obtained was left to settle down and starch was gotten by vacuum filtration.

Synthesis of silver nanoparticles (AgNps)
Sol-gel method described by Shahjahan (2017) was adopted with little modification. Briefly, beaker was thoroughly washed, after which 0.01 M prepared $AgNO_3$ and 0.1 M sodium hydroxide was added carefully to raise pH to 7. The solution was mixed rigorously for 45 min after which, (mass) cellulose or starch extracted from BSG was added as reducing agent. Black coloration shows that the solution has been reduced to silver nanoparticle. The change in colour shows the effect of surface plasmon resonance (SPR) of silver nanoparticle (Sathiya and Akiladeswari, 2014). The solution was continually stirred rigorously for another 3 hours as shining silver nanoparticles were formed over the solution and it was filtered, washed with deionized water and oven dried at 60ºC.

Characterization
UV–vis spectroscopy
The sample (1 g) of AgNps, AgNps-S and AgNps-C was diluted with 50 mL of distilled water. Ultra-violet visible spectrophotometer (Shimadzu uv-1650) was used to scan the sample between wavelengths of 300 to 600 nm and the optical band gap was determined using Tauc equation.

Tuac's Equation (as in Equation 1) was employed to determine band gaps for the samples.

$$\alpha h f = k(hf - E_g)^r \qquad (1)$$

Where α is absorbance coefficient; h is Planck constant; f is frequency; r is ½ for direct band gap; 2 for indirect band gap; 3/2 for forbidden direct band gap and 3 for forbidden indirect band gap. The absorbance coefficient was determined by equation 2.

$$\alpha = \frac{2.303A}{d} \qquad (2)$$

Where A is absorbance, d is the film thickness. This result in plotting $(\alpha h f)^2 against hf$ and extrapolate at point where peak of the absorption.

Fourier transform infrared (FTIR) Analysis

Functional group in silver nanoparticles and silver nanoparticles mixed with cellulose was determined using FT-IR spectroscopy (8400S, Shimadzu Scientific Instruments Inc.). Briefly, sample (1.0 µL) was placed on fused KBr disc. This was placed on cell holder, clamped loosely and fixed on the infrared (IR) beam. FTIR spectra were recorded in the range from 400 to 4000 cm^{-1}. The spectrum obtained was interpreted according to Willams (1982).

Scanning electron microscope (SEM)- Energy-dispersive X-ray spectrometer (EDS) analysis

AgNps and AgNps-C were scanned by VEGAB TESCAN Scanning Electron Microscope (SEM) equipped with BRUKER energy-dispersive X-ray spectrometer (EDS) for morphology and elemental characterization. The sample was pasted on the double sided cellotape placed on the SEM sample loader known as stub. The stub was mounted on stage after which chamber was tightly closed and the SEM was set to create vacuum as air in the chamber was evacuated. Higher HT voltage was not used since the sample is highly charged particle, hence 3.0 V was used.

Isolation and identification of microorganisms from fish and meat

Fish and meat were purchased from Obada market, Odeomu, Osun state in Nigeria. Fish and meat were separately homogenized. One gram was aseptically taken into 10 mL sterile distilled water to prepare stock. Serial dilution was carried out up to10^{-4} and 10^{-5} dilution factor. An aliquot of 0.1 mL was transferred into the petri dishes and the agar, nutrient agar, MacConkey agar, Baird Parker medium, Mannitol salt agar, Salmonella shigella agar, Eosin methylene blue agar, Violet Bile agar and Potato dextrose agar. The Petri dishes were incubated for 24 hours at 37° C for bacteria, while Potato Dextrose agar was incubated for 48 hours at 26°C for fungi. Biochemical tests such as catalase test, production of hydrogen sulphide (H$_2$S), indole, urease, methyl red, oxidase, coagulase, motility, methyl red, Voges-Proskauer, starch hydrolysis and sugars fermentation were carried out using the methods described by Cheesbrough (2006). Microorganisms with multiple antibiotic resistance were selected for further studies.

Antimicrobial activity of AgNps-C and AgNps-S

Antimicrobial activity of AgNps-C and AgNps-S were tested against microorganisms using agar well diffusion method (Cheesbrough, 2006). Distilled water was used as the negative control, amoxicillin and ketoconazole was used as positive control against bacteria and fungi, respectively. Bacteria were cultivated on nutrient broth at 37 ^{o}C for 24 h, while fungi were grown on potatoe dextrose broth at 25 ^{o}C for 48 h. Turbidity of inoculum was adjusted to 0.5 McFarland turbidity standard at 600nm using visible spectrophotometer. A sterile swab stick was moistened with bacterial and fungal inoculum and spread on Muller Hinton agar and Potato Dextrose agar, respectively. Afterwards, well of 4 mm diameter were bored into agar medium and filled with AgNps-C, AgNps-S and positive control. The plates were incubated at 37°C for 24 hours for bacteria and 25°C for 48-72 hours for fungi. Thereafter, zones of inhibition were measured in millimeter (mm) and recorded. Minimum inhibitory concentration (MIC) was was recorded as the lowest concentration that prevented the growth of microorganisms.

Evaluation of preservative effect of AgNps-C and AgNps-S on meat and fish

The preservative potential of AgNPs-c and AgNPs-s was assayed using the methods of Shavisi et al., (2017) and Badawy et al. (2019) with modification. Briefly, meat or fish (10 g) were aseptically weighed, boiled and placed in a sterile polyethylene bags with AgNps-C and AgNps-S. The samples were stored at -4 ^{o}C and another set at 37°C for 4 days. Microbial quality of preserved fish and meat was determined for 4 days.

Statistical analysis

Experiment was performed in replicate (n = 3). Data obtained were subjected to one-way analysis of variance (ANOVA). Values were presented as mean ± standard deviation (SD) using Statistical Package for Social Sciences (SPSS) version 23 software.

Results and discussion

Cellulose and starch from agro-waste (BSG) as a source of biomaterial

BSG is a potential source of polymers that can be successfully used for industrial and biotechnological applications. Finding of Mishra et al. (2017) revealed that BSG contains cellulose of 16 to 25% and as a potential source of cellulose nano-fibre that can be utilised for various purposes. Cellulose from orange peel waste and natural origin (other wastes) appears to be a good capping agent for the green synthesis of silver nanoparticles (Garza-Cervantes et al., 2020). Cellulose has gained much attention as bio-resourceful agent due to higher hydroxyls group in its molecular structure (Wu et al., 2012). Starch (C$_6$H$_{10}$O$_5$) n as a common carbohydrate and a polymer of glucose was extracted from BSG. The functional groups in biopolymer can act as reducing agent and as a capping agent to nanoparticles in order to demonstrate efficient bioactivities like antimicrobial, antioxidant, anticancer, and anti-inflammatory agents (Mohan et al., 2016). Therefore, biopolymers from renewable biomass is a promising and exciting biomaterial for biomedical applications because they abundantly available, renewability, no toxic, biocompatible, and easily biodegraded. Hence, biopolymers possess excellent mechanical properties that make them a good candidate for reinforcement materials in production of nanocomposites.

Characterization of synthesized nanoparticles and nanocomposites

AgNPs, AgNPs-C and AgNPs-S exhibit absorption at wavelength between 355 nm to 430 (Figure 1). This shows great effect of surface plasmon resonance exist within the range of 390 nm to 420. Studies of Lomelí-Marroquín *et al.* (2019) revealed 400 nm for AgNps. Band gap of 2.74 eV for Ag Nps, 2.72 eV for AgNps –S and 2.52 eV for AgNps –C was obtained in this study (Figure 2). AgNps –C has lowest band gap. The smaller the band gap, the easier for ionic interaction between the valence band and

conducting band. This shows that ions interchange was higher in AgNps-C than others, hence it will be effective than others. Huber et al (2012) revealed that bulk materials and thin films of conjugated polymers showed low optical band gaps.

Fourier Transform Infrared Spectroscopic Interpretation

The FTIR spectra of AgNps and AgNps-C are presented in Figure 3a and b. FTIR revealed various functional groups in biomolecules responsible for bio-reduction of Ag^+ and capping/stabilization of AgNps. The

wavelength of light absorbed is salient feature of chemical bonds in annotated spectrum. Tables 1 and 2 depicts data on peak values and probable functional groups obtained by FTIR from Figures 3a and b. Bands at 3425.69 cm^{-1} and 2926.11 cm^{-1} in the spectra corresponds to N-H stretch vibration, which are the primary and secondary amides (Phanjom and Ahmed, 2015), 3327.32 cm^{-1} signifies O–H vibration in cellulose, which helps in stabilizing silver ions into the cellulose fibers as results of interaction between cellulose hydrogen bonds and the metal nanoparticle and thus, makes the AgNps-C more efficient in formation of nano-composites (Musino et al., 2021).
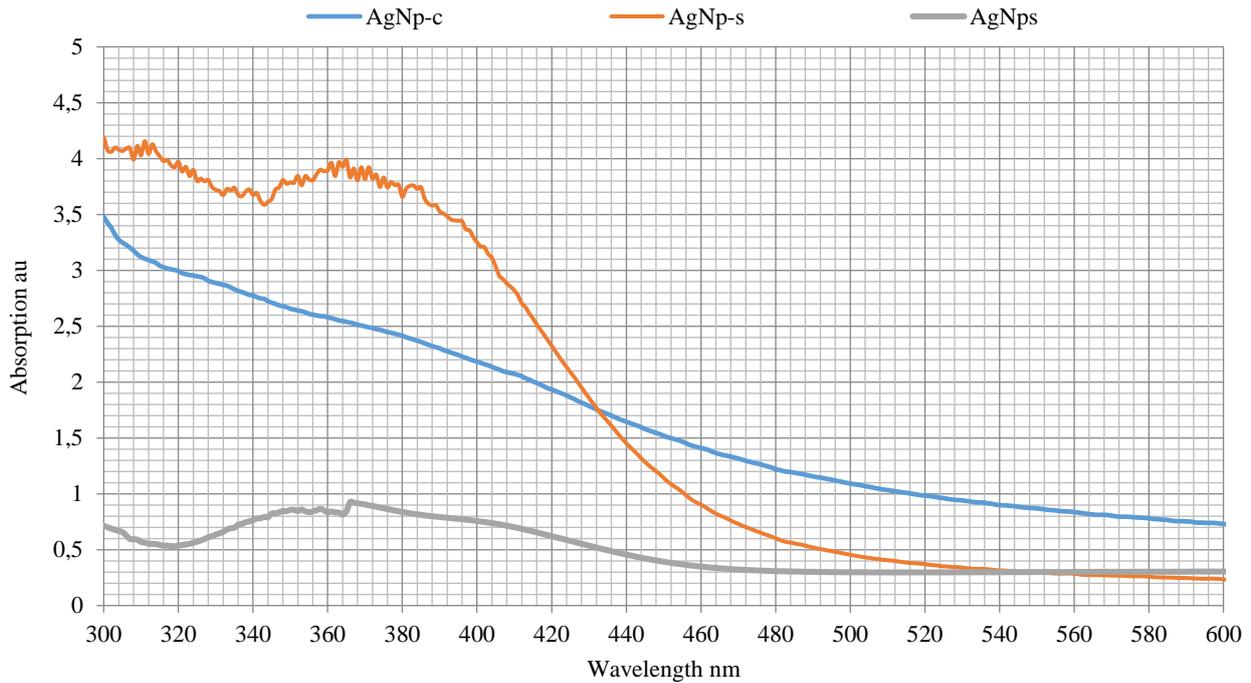

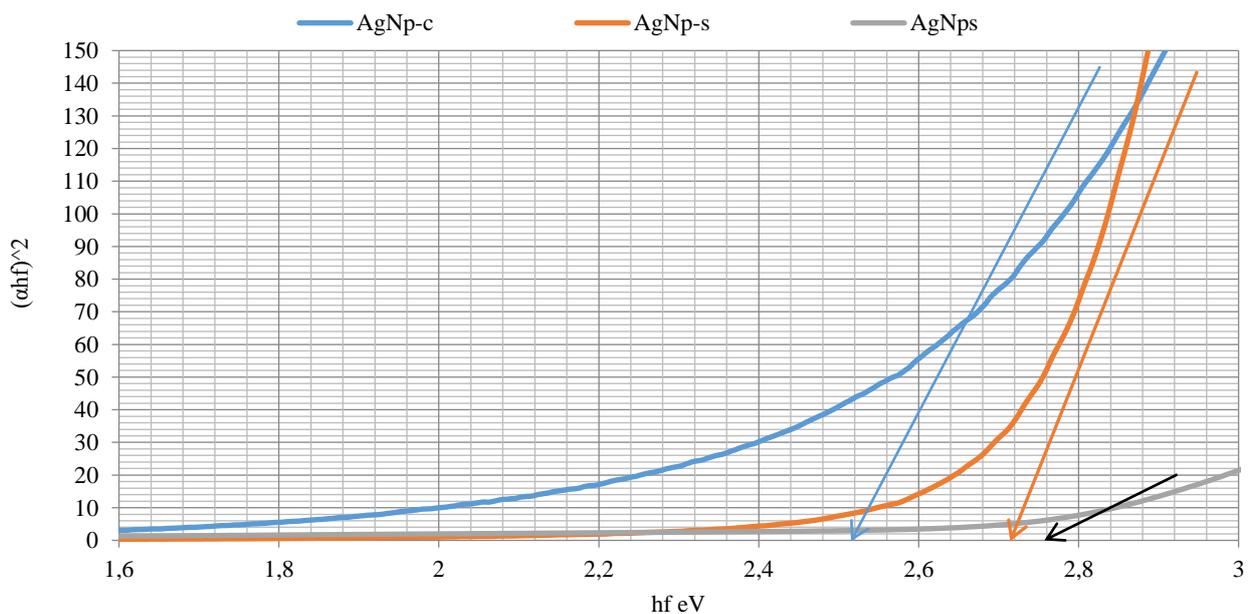Figure 1. UV-Vis absorption spectrum for AgNps, AgNps-C and AgNps-S

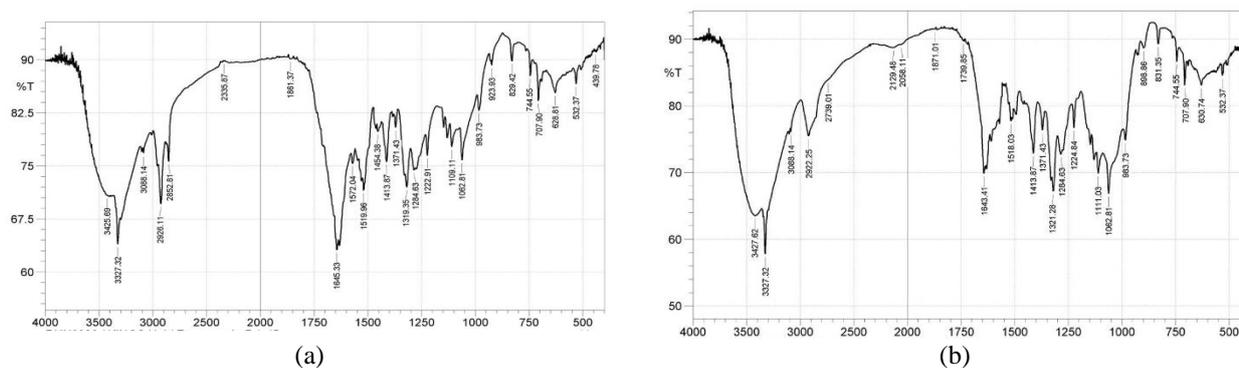Figure 2. Optical band gap for AgNps-C, AgNps-S and AgNps

(a)

(b)

Figure 3. FTIR spectra of (a) AgNps and (b) AgNps -C

Table 1. FTIR spectra peak and functional groups in AgNps

Peaks wavelength (cm^{-1})	Functional groups
532.37	Silver
620.81	CH out of plane bending of Methylene group
1062.81	N-H In-plane bending
1284.63	C-O Stretch
1454.38	N-H Stretch
1572.04	N-H bend
1645.33	Amide functional group
2852.81	Aromatic –C-H stretch
2926.11	N-H Stretch
3088.14	C=H Stretch Aromatics
3327.32	O-H stretch
3425.69	N-H Stretch

Table 2. FTIR spectral peak and functional groups in AgNps-C

Peaks wavelength (cm^{-1})	Functional groups
532.37	Silver
630.74	CH out of plane bending of Methylene group
1082.81	N-H In-plane bending
1284.83	C-O Stretch
1518.03	C-C Stretch in aromatics
1643.41	Amide functional group
1739.85	C=O stretch of aldehyde
2129.48	Methylene group
2739.01	Aldehyde C-H stretch
2922.25	N-H Stretch
3088.14	C=H Stretch Aromatics
3327.32	O-H stretch
3427.62	N-H Stretch

Band at 2852.81 cm^{-1} region arising from C–H stretching of aromatic compound were observed, the band at 1645.33 cm^{-1} in the spectra corresponds to amide functional group arising due to carbonyl stretch (Prakash et al., 2013). Peak at 1572. 04 cm^{-1} corresponds to N-H bend, the band at 1454.38 cm^{-1} was assigned for N–H stretch vibration present in amide linkages (Phanjom and Ahmed, 2015). Peak at 1284.63 cm^{-1} refer to C-O stretching vibrations mode. The carbonyl groups have strong ability to bind to silver (Balaji et al., 2009). The bands at 1062.81 cm^{-1} are assigned for N–H stretch vibration. The peak at 829.42 cm^{-1} assigned to C=CH$_2$ and peaks near 628.81 cm^{-1} assigned to CH out of plane bending vibrations are substituted ethylene systems –CH=CH (cis) (Priya et al., 2011). Some bands in Fig. 3b are similar to Fig. 3a, verifying the functional groups, there is a shift in some bands due to the presence of cellulose in the blend.

Some bands came up; 2739.01 cm^{-1} indicates C-H stretch of aldehyde, 2129.48 cm^{-1} show the methylene group, this is similar to the findings of Priya et al. (2011), 1739.85 cm^{-1} representing aldehydric CO stretch. This showed that cellulose was oxidized by reduction of silver ion and the alkanol group oxidized to aldehyde group (Hameed et al., 2017).

SEM-EDX

The SEM morphology at different magnification is shown in Figures 4 to 6. The particle of AgNps shows spherical shape with particulate size ranged from 6.38 to 53.43 nm. Findings of Ibrahim (2015) indicated spherical shape for silver nanoparticles. The diameter obtained in this study is within the range of nanoparticle (0-100 nm), which agreed the findings of Balashanmugam et al. (2013) who reported 20-50 nm for silver nanoparticle.
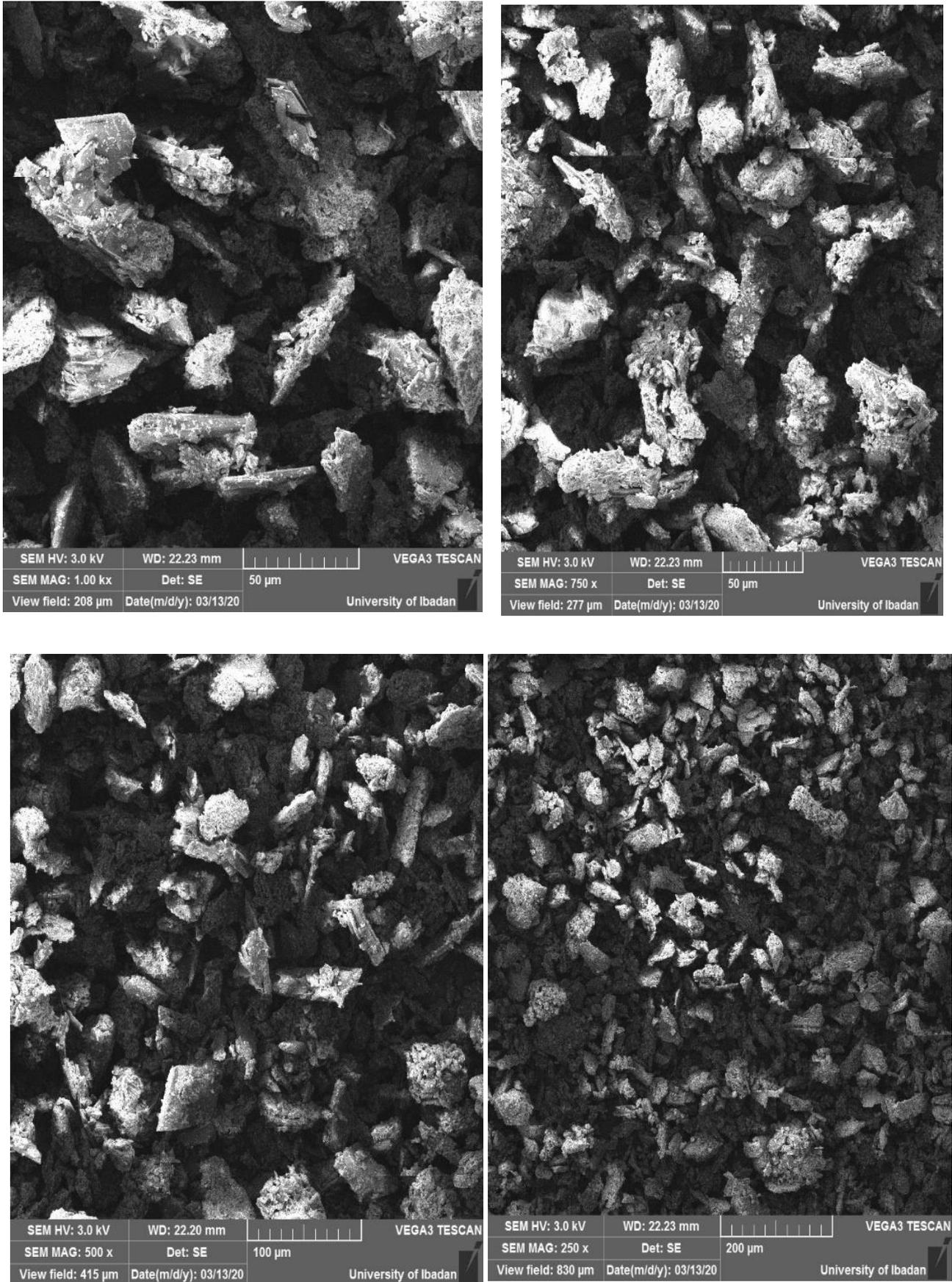
Figure 4. SEM at different magnification ranged from 1000 to 250 at 3. 0 KV for AgNps
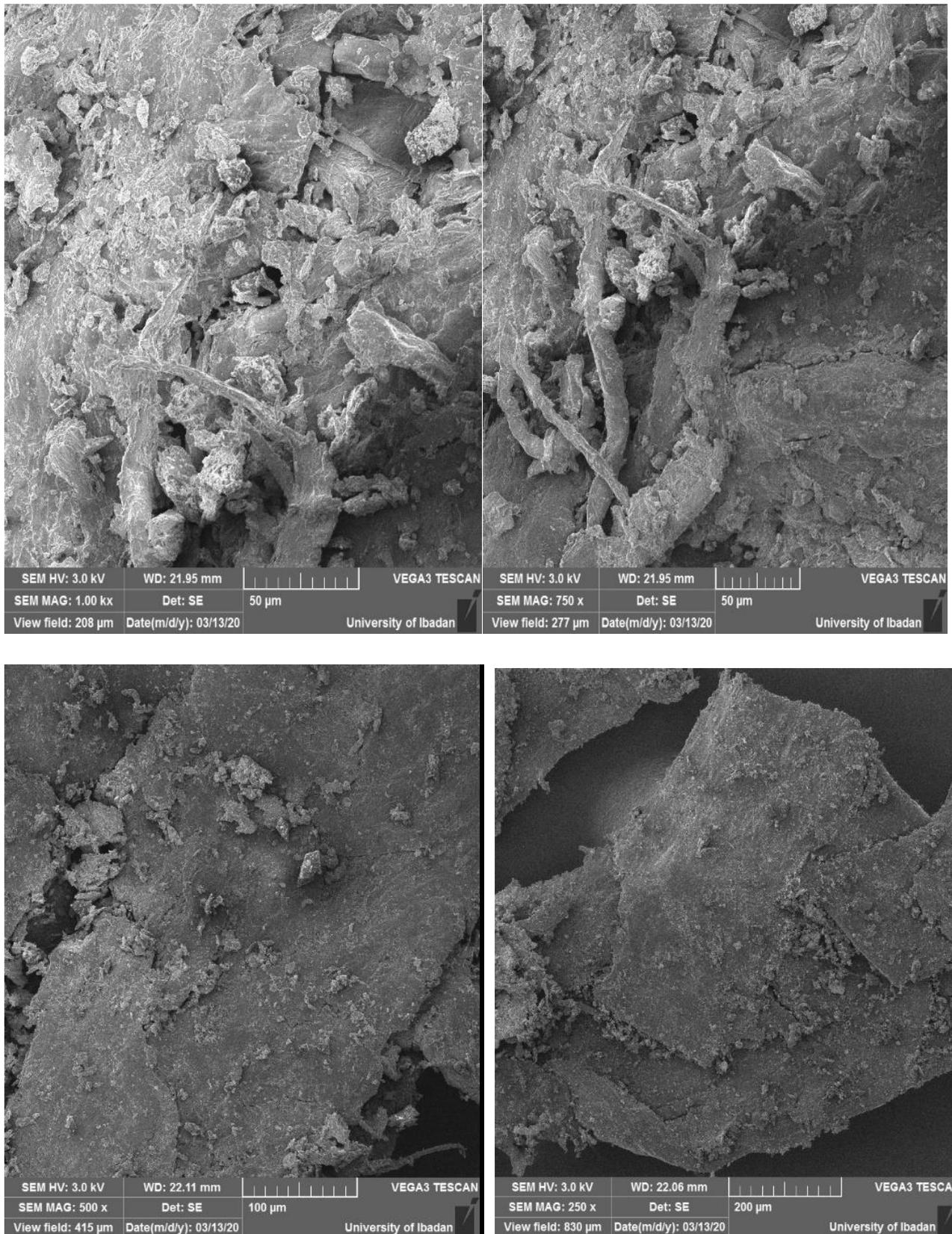
Figure 5. SEM images at different magnification ranged from 1000 to 250 at 3. 0 KV for AgNps-C
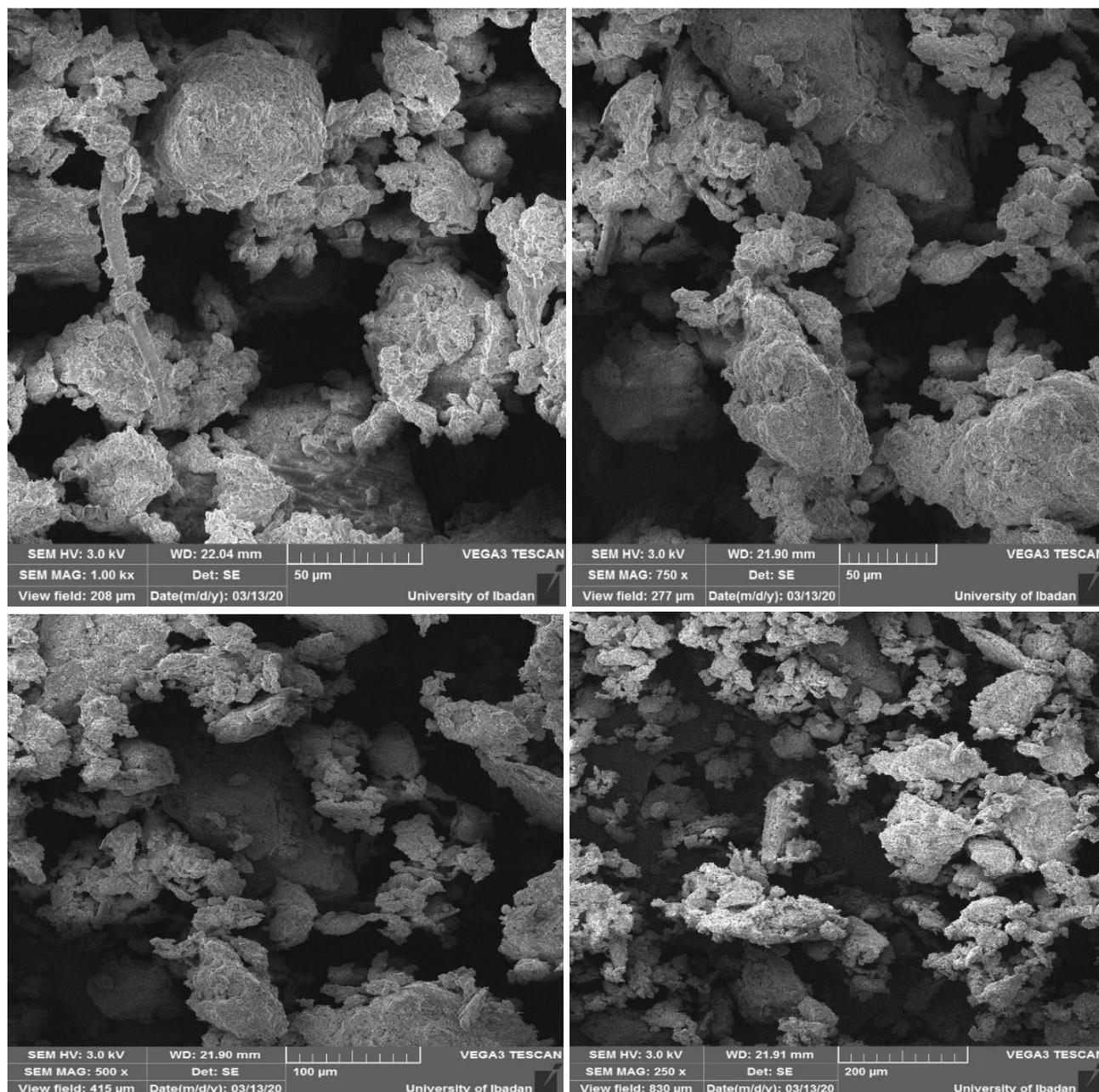
Figure 6. SEM at different magnification ranged from 1000 to 250 at 3. 0 KV for AgNps-S

The cellulose and starch from BSG characterized with the SEM show fiber structure with spherical shapes (Figures 5 and 6) which follows the findings of Liew et al. (2015) who extracted cellulose from bamboo. The particle shows fiber elliptical shape with particulate size ranged from 6.97 to 128.77 nm and 4.80 to 20.81 nm for AgNps-S and AgNps-C respectively. EDS of AgNps shows 87.01 % of silver with 7.58 % of carbon and 5.41 % of oxygen (Figure 7). The carbon was owned to the constituent of cellulose from BSG used as a reducing agent. The EDS of AgNps-C shows 76.49 % of carbon, 16.01 % of oxygen, 3.87 % of silver, 2.90 % of sodium and 0.67 % of phosphorus as shown in Figure 7b. The higher proportion of carbon in AgNps-C is as a result of mixing ratio of cellulose to AgNps (10:1). The presence of sodium and phosphorus could be attributed to composition of BSG. The report of Muthusamy (2014) and Silbir and Goksungur (2019) revealed the BSG contains phosphorus, sodium and other minerals. The EDS shows the binding energy for AgNPs at 0.330 KeV L-series based on the lower activation energy used to dispersed the nanoparticle as shown in Figure 7 a and b. This is similar to the findings of Kumr et al. (2018) who spotted binding energy of AgNPs at 0.33 KeV. Studies of Balashanmugam et al. (2013) and Alexander et al. (2018) have obtained the binding energies of Ag 3d5/2 and Ag 3d3/2 peaks 367.73 and 373.71 eV correspond to Ag_2O state.

Antimicrobial and preservative potentials of AgNps-C and AgNps-S

Table 3 shows zones of inhibition displayed by AgNps-C and AgNps-S against microorganisms isolated from meat and fish. AgNps-C displayed inhibited zones of 6.11 mm to 12.40 mm and 9.04 mm to 13.30 mm against microorganisms isolated from fish and meat, respectively. AgNps-S displayed inhibitory zones of 5.00 mm to 10.30 mm against indicator microorganisms.
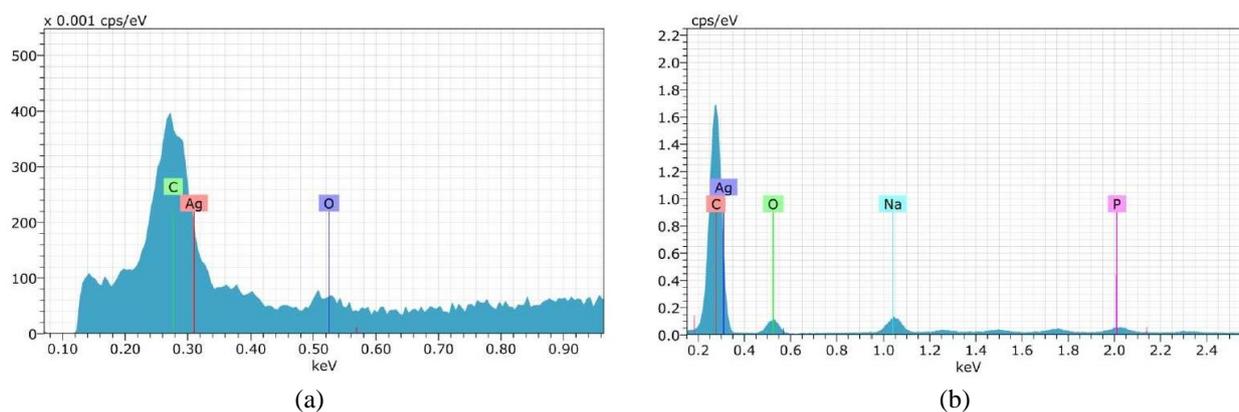
(a)　　　　　　　　　　　　　　　　　　　　(b)

Figure 7. EDS of (a) AgNps and (b) AgNps-C

Table 3. Zones of inhibition displayed by AgNp-C and AgNp-S at 1000 μg/mL

Source	Microorganisms	AgNp-C	AgNp-S	Amoxicillin/ ketoconazole
Fish	*E. coli*	12.40±0.02	10.30±0.01	17.30±1.20
	P. aeruginosa	6.11±0.00	0.0	12.2±1.64
	P. mirabilis	8.00±0.10	6.00±0.00	10.10±0.00
	K. pneumoniae	8.00±0.30	0.0	8.00±0.01
	K. oxytoca	10.00±0.00	7.00±0.03	12.40±0.02
	C. albicans	9.03±0.20	5.00±0.00	11.30±0.06
Meat	*Bacillus subtilis*	12.00±1.20	8.00±0.40	15.30±2.01
	Streptococcus faecalis	9.04±0.52	0.0	12.00±1.05
	Micrococcus luteus	11.00±0.40	6.00±0.01	12.10±1.50
	P. aeruginosa	10.30±0.62	0.0	16.10±1.20
	S. aureus	9.00±0.30	6.02±0.01	8.00±0.50
	E. coli	13.30±1.00	0.0	13.00±0.92
	K. pneumoniae	13.00±0.72	8.04±0.33	14.05±0.82
	[#]*E. coli*	11.00±0.61	0.0	12.10±0.73
	[#]*S. aureus*	12.06±1.04	7.00±0.01	10.00±0.62
	[#]*C. albicans*	11.01±0.90	6.00±0.04	14.03±1.20

[#] clinical isolates with multiple drug resistance. 0.0= no zones of inhibition observed at 1000 μg/mL

Table 4. MIC (μg/mL) of AgNps-C and AgNps-S against tested microorganisms

Source	Microorganisms	AgNps-C	AgNps-S
Fish	*E. coli*	125	500
	P. aeruginosa	250	0.0
	P. mirabilis	250	1000
	K. pneumoniae	125	0.0
	K. oxytoca	500	1000
	C. albicans	500	500
Meat	*Bacillus subtilis*	250	500
	Streptococcus faecalis	500	0.0
	Micrococcus luteus	125	1000
	P. aeruginosa	500	0.0
	S. aureus	125	1000
	E. coli	125	0.0
	K. pneumoniae	250	1000
	[#]*E. coli*	125	0.0
	[#]*Staphylococcus aureus*	125	500
	[#]*C. albicans*	500	1000

[#] clinical isolates with multiple drug resistance. 0.0 = MIC > 1000 μg/mL

Findings of Almeida *et al*. (2015) revealed that *Escherichia coli, Enterococcus faecalis, Staphylococcus aureus, S. epidermidis, Vibrio cholerae, Pseudomonas aeruginosa, Shigella flexneri, Bacillus anthracis, Vibrio cholerae, Pseudomonas aeruginosa, B. subtilis, B. cereus, Proteus mirabilis, Salmonella enterica typhimurium, Micrococcus luteus, Listeria monocytogenes*, and *Klebsiella pneumonia* were inhibited using nanoparticles. Yakout and Mostafa (2015) ascertained that synthesized green AgNps with soluble starch showed a pronounced antibacterial activity with zones of inhibition ranged from 7.2 mm to 16. 3 against Grams positive and negative pathogenic bacteria like *Staphylococcus aureus* and *Streptococcus pyogenes, Salmonella typhi, Shigella sonnei*

and *Pseudomonas aeruginosa*. In this study, AgNps-C exhibited better inhibitory potential than AgNps-S. Silver nanoparticles (AgNps)-cellulose exhibit good biocompatibility and antimicrobial properties because AgNps are tightly immobilized on cellulosic materials and bestowing better antibacterial activity (Xu et al., 2018). Silver nanoparticle-cellulose as bio-composite inhibited the growth of *Escherichia coli* ATCC 11229, *Staphylococcus aureus* ATCC 6538, *Pseudomonas aeruginosa* ATCC 27853, multidrug resistant *Staphylococcus aureus* and *Pseudomonas aeruginosa* (Garza-Cervantes et al., 2020). In the findings of Xu et al. (2019), silver nanoparticles (AgNps) with dialdehyde cellulose nanocrystal exhibited antibacterial activities against *Escherichia coli* and *Staphylococcus aureus*, which later added to pulp fibers to obtain hand sheets with strong antibacterial activities. Thiagamani et al. (2019) revealed that infusion of AgNps on cellulose-based hybrid nanocomposites maintain its mechanical, thermal and proved to have good antimicrobial activities, which therefore, recommended for active packaging applications. Rozilah et al. (2020) revealed antibacterial activity of nano-composite films containing sugar palm starch and sugar palm nanoparticles crystalline cellulose with inhibited zones ranged from 0.1 mm to 8.0 mm against the growth of *Escherichia coli*, *Staphylococcus aureus*, and *Salmonella cholerasuis*. Silver nanoparticles and silver nanocomposites have a wide spectrum of antimicrobial activity with different mechanism of action against Gram-positive and negative bacteria, fungi, and viruses. Studies of Morones *et al.* (2005); Sobye *et al.* (2015); and Simbine et al. (2019) have revealed that nanoparticles in the range

of 1 to 10 nm bound to the surface of the cell membrane and drastically disrupt its permeability and respiration, and possibly interact with macromolecules in cells such as proteins, deoxyribonucleic acid (DNA). This reduce the growth and metabolism of bacterial cells and therefore, leading to accelerated lysis (Sobye *et al.*, 2015).

Table 4 shows the minimum inhibitory concentrations (MIC) of AgNps-C and AgNps-S against tested microorganisms. The MIC ranged from 125 µg/mL to 500 µg/mL and 500 µg/mL to 1000 µg/mL for AgNps-C and AgNps-S, respectively. Finding of Badawy et al. (2019) obtained MIC of 200 and 250 mg/L against *E. coli* and *Salmonella typhimurium*, respectively when chitosan-silver nanoparticles used in preservation of minced meat. Table 5 shows the preservative quality of AgNps-C and AgNps-S on fish and meat at different temperature for 4 days. The microbial load obtained on each sample; fish and meat were recorded. At day 1- 3, the microbial load ($\times 10^5$ cfu/g) of fish and meat decreased from 1.4 to 1.0 and 1.5 to 1.3 at 4 °C when treated with AgNps-C. AgNps-S did not significantly reduced the microbial load but maintained microbial load of 1.9 -2.0 and 2.0-2.3 $\times 10^5$ cfu/g on fish and meat respectively. At day 4, the untreated fish and meat have higher microbial load with traces of spoilage at day 4. Findings of Badawy et al. (2019) revealed lower microbial load of 8.17 to 14.47 $\times 10^4$ cfu/g when chitosan-silver nanoparticles was used to preserve minced meat. In the study of Shavisi et al. (2017), microbial population decreased to 1-3logCFU/g in treated minced beef with extended shelf life without any unfavourable organoleptic properties when cellulose nanoparticle was combined with *Ziziphora clinopodioides* essential oil.

Table 5. Microbial quality ($\times 10^5$ cfu/g) of preserved fish and meat with AgNps-C and AgNps-S at different storage conditions

Day(s)	1			2		
	AgNps-C	AgNps-S	NT	AgNps-C	AgNps-S	NT
Fish at 4 °C	1.4	1.9	3.8	1.0	1.8	5.4
Meat at 4 °C	1.5	2.0	3.7	1.3	1.8	5.6
Fish at 29 °C	4.4	10.6	14.4	4.6	6.7	18.5
Meat at 29 °C	4.9	12.2	16.0	5.0	7.1	19.2
Day(s)	3			4		
	AgNps-C	AgNps-S	NT	AgNps-C	AgNps-S	NT
Fish at 4 °C	1.0	2.0	7.2	1.6	2.7	8.6
Meat at 4 °C	1.3	2.3	7.6	1.8	3.0	8.8
Fish at 29 °C	4.9	7.9	sp	6.4	8.0	sp
Meat at 29 °C	5.6	9.3	sp	7.5	8.6	sp

NT: sample was not treated with AgNps-C or AgNps-S; SP: Samples were smelling and spoiled

Conclusion

Polymers should be incorporated with silver nanoparticles because of their antimicrobial properties against pathogenic organisms which will also go a long way to reduce environmental pollution in our environments. The application of silver nanoparticles (AgNps) with biopolymer from agrowastes for food packages will offer new perspectives to prevent microbial spoilage and increase the shelf life of foods. AgNps can be incorporated to non-degradable (polyethylene, polyvinyl chloride, vinyl alcohol) and biodegradable polymers such as cellulose, starch, chitosan, agarose) to produce food packages.

Declaration section

Statement of ethical approval
Compliance with ethical standards
Conflict of interest
All authors declare no conflict of interest.

References

Almeida ACS, Franco AE N, Peixoto FM, Pessanha KL, Melo NR. 2015. Application of nanotechnology in food packaging. Polímeros, 25: 89-97. http://dx.doi.org/10.1590/0104-1428.2069.

Araújo DJC, Machado AV, Vilarinho MCLG. 2019. Availability and Suitability of Agroindustrial Residues as Feedstock for Cellulose-Based Materials: Brazil Case Study. Waste and Biomass Valorization 10: 2863–2878 https://doi.org/10.1007/s12649-018-0291-0.

Badawy MEI, Lotfy TMR, Shawir SMS. 2019. Preparation and antibacterial activity of chitosan-silver nanoparticles for application in preservation of minced meat. Bull Natl Res Cent 43, 83 https://doi.org/10.1186/s42269-019-0124-8.

Bajpai VK, Kamle M, Shukla S, Mahato DK, Chandra P, Hwang SK, Kumar P, Huh YS, Han YK. 2018. Prospects of using nanotechnology for food preservation, safety, and security. Journal of food and drug analysis, 26(4), 1201–1214. https://doi.org/10.1016/j.jfda.2018.06.011

Balaji S., Basavaraja R, Deshpande DB, Mahesh BK, Prabhakar A, Venkataraman, (2009). Colloid Surf. B: Biointerf. 68 - 88.

Balashanmugam P, Santhosh S, Giyaullah H, Balakumaran MD, Kalaichelvan PT. 2013. Mycosynthesis, Characterization and Antibacterial Activity of Silver Nanoparticles from *Microporus xanthopus*: A Macro Mushroom. International Journal of Innovative Research in Science, Engineering and Technology 2(11): 6262-6270.

Bodunde RS, Ogidi CO, Akinyele BJ. 2019. Load and antibiotic susceptibility pattern of microorganisms in muscle foods sold in Akure, Southwest Nigeria. Journal of Food Quality and Hazards Control 6: 30-36. https://doi.org/10.18502/jfqhc.6.1.456

Cheesbrough M. 2006. District laboratory practice for tropical countries, part 2. 2nd ed. Cambridge, UK. 62-70, 267-330.

Das MP, Livingstone JR, Veluswamy P, Das J. 2017. Exploration of *Wedelia chinensis* leaf-assisted silver nanoparticles for antioxidant, antibacterial and *in vitro* cytotoxic applications. J Food Drug Anal. https://doi.org/10.1016/j.jfda.2017.07.014.

Lomelí-Marroquín D, Medina Cruz D, Nieto-Argüello A, Vernet Crua A, Chen J, Torres-Castro A, Webster TJ, Cholula-Díaz JL. 2019. Starch-mediated synthesis of mono- and bimetallic silver/gold nanoparticles as antimicrobial and anticancer agents. Int J Nanomedicine, 27, 14:2171-2190. doi: 10.2147/IJN.S192757.

Duncan TV. 2011. Applications of nanotechnology in food packaging and food safety: barrier materials, antimicrobials and sensors. Journal of Colloid and Interface Science, 363(1): 1–24. https://doi.org/10.1016/j.jcis.2011.07.017.

Fariq A, Khan T, Yasmin A. 2017. Microbial synthesis of nanoparticles and their potential applications in biomedicine. Journal of Applied Biomedicine. 15(4): 241-248.

Garza-Cervantes JA, Mendiola-Garza G, de Melo EM. et al. Antimicrobial activity of a silver-microfibrillated cellulose biocomposite against susceptible and resistant bacteria. Sci Rep 10, 7281 (2020). https://doi.org/10.1038/s41598-020-64127-9.

Al-Hajjar HA, Oda AM, Rasheed MH, Naj HK, Hilwas MH. 2017. Synthesis and characterization of silver nanoparticles loaded with cellulose and cellulose carbamate. Journal of Chemical and Pharmaceutical Research, 9(12):98-103.

He X, Hwang HM. 2016. Nanotechnology in food science: functionality, applicability, and safety assessment. Journal of food and drug analysis, 24(4), 671–681. https://doi.org/10.1016/j.jfda.2016.06.001.

Huber J, Jung C, Mecking S. 2012. Nanoparticles of Low Optical Band Gap Conjugated Polymers. Macromolecules, 45, 19, 7799–7805

Ibrahim HMM. 2015. Green synthesis and characterization of silver nanoparticles using banana peel extract and their antimicrobial activity against representative microorganisms. Journal of Radiation Research and Applied Sciences, 1 -11.

Israel AU, Obot IB, Umoren SA, Mkepenie V, Asuquo JE. (2008). Production of Cellulosic Polymers from Agrowastess. E-Journal of Chemistry. 5(1):81-85.

Liew FK, Hamdan S, Rahman MR, Rusop M, Lai JCH, Hossen F, Rahman MM. 2015. Synthesis and Characterization of Cellulose from Green Bamboo by Chemical Treatment with Mechanical Process. HPCJC, (2015) 6-13.

Lorenzo JM, Munekata PE, Dominguez R, Pateiro M, Saraiva JA, Franco D. 2018. Main Groups of Microorganisms of Relevance for Food Safety and Stability: General Aspects and Overall Description. Innovative Technologies for Food Preservation, 53–107. https://doi.org/10.1016/B978-0-12-811031-7.00003-0

Maraveas C. 2020. Production of sustainable and biodegradable polymers from agricultural waste. Polymers, 12(5), 1127; https://doi.org/10.3390/polym12051127

Mishra PK, Gregor T, Wimmer R. 2017. Utilising brewer's spent grain as a source of cellulose nanofibres following separation of protein-based biomass, BioRes. 12(1), 107-116.

Mohan S, Oluwafemi OS, Kalarikkal N, Thomas S, Songca SP. 2016. Biopolymers – application in nanoscience and nanotechnology. doi: 10.5772/62225.

Morones JR, Elechiguerra JL, Camacho A, Holt K, Kouri JB, Ramírez JT, Yacaman MJ. 2005. The bactericidal effect of silver nanoparticles. Nanotechnology, 16(10):2346-2353. http://dx.doi.org/10.1088/0957-4484/16/10/059.

Motaung TE. Linganiso LZ. 2018 Critical review on agrowaste cellulose applications for biopolymers. International Journal of Plastics Technology, 22(2):185–216 https://doi.org/10.1007/s12588-018-9219-6

Motelica L, Ficai D, Ficai A, Oprea OC, Kaya DA. Andronescu E. 2020. Biodegradable Antimicrobial Food Packaging: Trends and Perspectives Foods, 9, 1438; doi:10.3390/foods9101438

Musino D, Rivard C, Landrot G, Novales B, Rabilloud T, Capron I. 2021. Hydroxyl groups on cellulose nanocrystal surfaces form nucleation points for silver nanoparticles of varying shapes and sizes, Journal of Colloid and Interface Science, 584, 360-371.

Muthusamy N 2014. Chemical composition of brewers spent grain-A review. International Journal of Science, Environment and Technology 3(6): 2109-2112

Nevárez LM, Casarrubias LB, Canto OS, Celzard A, Fierro V, Gómez RI, Sánchez GG. 2011 Biopolymers-based nanocomposites: membranes from propionated lignin and cellulose for water purifcation. Carbohydr Polym 86:732–741.

Pornchai R. 2009. Blended Films of Carboxymethyl Cellulose from Papaya Peel (CMCp) and Corn Starch Kasetsart J. Nat. Sci. 43: 259 - 266

Prakash P, Gnanaprakasam P, Emmanuel R, Arokiyaraj S, Saravanan M. 2013. Green synthesis of silver nanoparticles from leaf extract of *Mimusops elengi*, Linn. for enhanced antibacterial activity against multi drug resistant clinical isolates. Colloids and Surfaces B: Biointerfaces, 108: 255-259.

Preetha D, Prachi K, Chirom A, Arun R. 2013. Synthesis and Characterization of Silver Nanoparticles Using Cannonball Leaves and Their Cytotoxic Activity against MCF-7 Cell Line. https://doi.org/10.1155/2013/598328.

Priya AM, Selvan RK, Senthilkumar B, Satheeshkumar MK, Sanjeeviraja C. 2011. Synthesis and characterization of $CdWO_4$ nanocrystals. Ceramics International, 37, 7:2485–2488.

Phanjom P, Ahmed G. 2015. Biosynthesis of Silver Nanoparticles by *Aspergillus oryzae* (MTCC No. 1846) and Its Characterizations nanoscience and nanotechnology, 5(1): 14-20

Rozilah A, Aiza -Jaafar CN, Sapuan SM, Zainol I, Ilyas RA. 2020. The effects of silver nanoparticles compositions on the mechanical, physiochemical, antibacterial, and morphology properties of sugar palm starch biocomposites for antibacterial coating. Polymers, 12, 2605; doi:10.3390/polym12112605.

Saini JK, Saini R, Tewari L. 2015. Lignocellulosic agriculture wastes as biomass feedstocks for second-generation bioethanol production: concepts and recent developments. 3 Biotech, 5(4), 337–353. https://doi.org/10.1007/s13205-014-0246-5

Sathiya CK, Akilandeswari, S. 2014. Fabrication and Characterization of Silver nanoparticles using Delonix elata leaf broth. Spectrochimica acta Part A: Molecular and Biomolecular Spectroscopy 128 (2014) 337-341.

Shahjahan M, Rahman MH, Hossain MS, Khatun MA, Islam A, Begum MHA. 2017. Synthesis and Characterization of Silver Nanoparticles by Sol-Gel Technique. Science Publication Group, Nanoscience and Nanometrology; 3(1): 34-39.

Shavisi N, Khanjari A, Basti AA, Misaghi A, Shahbazi Y. 2017. Effect of PLA films containing propolis ethanolic extract, cellulose nanoparticle and *Ziziphora clinopodioides* essential oil on chemical, microbial and sensory properties of minced beef. Meat Science, 124, 95–104. https://doi.org/10.1016/j.meatsci.2016.10.015

Silbir S, Goksungur Y. 2019. Natural Red Pigment Production by Monascus Purpureus in Submerged Fermentation Systems Using a Food Industry Waste: Brewer's Spent Grain. Foods, 8(5), 161. doi:10.3390/foods8050161

Simbine EO, Rodrigues LC, Lapa-Guimarães J, Kamimura ES, Corassin CH, Oliveira, CAF. 2019. Application of silver nanoparticles in food packages: a review. Food Science and Technology, 39(4), 793-802..https://doi.org/10.1590/fst.36318.

Sobye A, Kolding A, Jorgensen JK, Lund MK, Mikkelsen MO. 2015. Bactericidal effect of silver nanoparticles: determination of size and shape of triangular silver nanoprisms and spherical silver nanoparticles and their bactericidal effect against Escherichia coli and Bacillus subtilis (pp. 9-76). Aalborg: School of Engineering and Science Nanotechnology

Srividya N, Ghoora MD, Padmanabh PR. 2017. Antimicrobial nanotechnology: research implications and prospects in food safety, Grumezescu, A. M. (Ed): In Nanotechnology in the Agri-Food Industry, Food Preservation, Academic Press, 125-165.

Temesgen S, Rennert M, Tesfaye T, Nase M. 2021. Review on Spinning of Biopolymer Fibers from Starch. Polymers, 13, 1121. https://doi.org/10.3390/polym13071121

Thiagamani SMK, Rajini N, Siengchin S, Rajulu AV, Hariram N, Ayrilmis N. 2019. Influence of silver nanoparticles on the mechanical, thermal and antimicrobial properties of cellulose-based hybrid nanocomposites. Composites Part B: Engineering, 165:516-525.

Thiago RDSM, Pedro PMDM, Eliana FCS. 2014. Solid wastes in brewing process: a review. Journal of Brew. Distill, 5:1-9.

Willams DHA. 1982. Spectroscopic method in organic chemistry. Willey and son, London.

Wu J, Ning Z, Zhang X, Jian X. 2012. Cellulose/silver nanoparticles composite microspheres: Eco-friendly synthesis and catalytic application. Cellulose, 19, 1239–1249.

Xu Q, Jin L, Wang Y. et al. 2019. Synthesis of silver nanoparticles using dialdehyde cellulose nanocrystal as a multi-functional agent and application to antibacterial paper. Cellulose 26: 1309–1321 https://doi.org/10.1007/s10570-018-2118-3.

Xu Y, Li S, Yue X, Lu W. 2018. Review of silver nanoparticles (AgNPs)-cellulose antibacterial composites, BioRes. 13: 2150-2170.

Yakout SM, Mostafa AA. 2015. A novel green synthesis of silver nanoparticles using soluble starch and its antibacterial activity. International journal of clinical and experimental medicine, 8(3), 3538–3544.

Zorraquín-Peña I, Cueva C, Bartolomé B, Moreno-Arribas MV. 2020. Silver Nanoparticles against Foodborne Bacteria. Effects at Intestinal Level and Health Limitations. Microorganisms, 8(1), 132. https://doi.org/10.3390/microorganisms8010132.