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Mechanical and antibacterial properties of recycled carton paper coated by PS/Ag nanocomposites for packaging

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ABSTRACT

Polymer nanocomposites and paper constitute a new class of packaging materials. In this study silver nanoparticles were prepared by novel method as antibacterial additive, where, synthetic takes place with aid of a novel, non-toxic, and eco-friendly biological materials namely rice straw (RS) powder. The prepared Ag nanoparticle was examined by transmission electron microscope (TEM), X-ray diffraction pattern (XRD) and UV-spectroscopy. The silver nanoparticles were then embedded into commercial polystyrene solution. The recycled carton paper was coated by the polystyrene nanocomposites containing different concentration of silver nanoparticles, namely 2, 4, 6 and 8% based on polystyrene. The prepared recycled carton sheets were characterized by scanning electron microscope (SEM). The mechanical properties, water vapor permeability and antibacterial effect of recycled carton sheets were also investigated.

1. Introduction:

Nanotechnology is presently recognized as one of the most promising areas for packaging technological improvement in the 21st century (Hu & Fu, 2003). Moreover, nanocomposites materials have become a rapidly expanding area of research, which cover an unlimited variety of systems with potentially novel material properties. When compared to pure polymers, polymer nanocomposites possess many attractive properties, such as enhanced barrier characteristics (Willett, 1994), increased moduli and strengths (Cho, Woo, Chun, & Park, 2001), high heat distortion temperatures, reduced gas permeabilities (Jong & Perry, 2007), and decreased absorption in organic liquids (Jaserg, Swanson, Nelsen, & Doane, 1992). While a large number of polymer nanocomposites systems have been studied, a great deal of attention has also been focused on polystyrene (PS) nanocomposites (Essawy, Badran, Youssef, & Abd El-Hakim, 2004a; Essawy, Badran, Youssef, & Abd El-Hakim, 2004b; Noh et al., 2000; Wang, Hao, Zhu, & Wilkie, 2002) as studied using cone calorimetry as well as thermogravimetric analysis (TGA). Also, the improvements in physical properties are obtained at very low of filler loadings according to Alexander and Dubois (2000) and Giannelis (1996). Furthermore, PS is a thermoplastic resin with good processing properties, where it is used in many applications including food packaging, domestic appliances, electronic goods, toys, household goods and furniture.

Paper is reported to be the most widely used material in packaging applications owing to its characteristics of printability, recyclability and biodegradability. However, since paper is hygroscopic and porous, its barrier properties against water-vapor, gases and aromas are poor. To improve its barrier properties, paper is often impregnated with coatings from synthetic polymers such as polystyrene, polyethylene, polyvinyl alcohol, rubber latex and fluorocarbon. Theses polymers fill the paper pores and form a dense layer at the paper surface (An, Zhang, Wang, & Tang, 2008; Mbhele et al., 2003). An antimicrobial nanocomposites film is particularly desirable due to its acceptable structural integrity and barrier properties imparted by the nanocomposites matrix, and the antimicrobial properties contributed by the natural antimicrobial agents impregnated within polymer matrix (Haroun & Youssef, 2011; Jong & Perry, 2007). The most common nanocomposites used as antimicrobial films for food packaging are based on silver, which is well known for its strong toxicity to a wide range of microorganisms (Liau, Read, Pugh, Furr, & Russell, 1997), with high temperature stability and low volatility (Kumar & Munstedt, 2005). Furthermore, silver nanoparticles can be synthesized by several physical, chemical and biological methods (Nair & Laurencin, 2007; Sharma, Yngard, & Lin, 2009; Zhang, Peng, Huang, Zhou, & Yan, 2008). Naturally available agricultural wastes such as banana peels that are inherently rich in polymers such as lignin, hemicellulose and pectins have been investigated for the synthesis of silver nanoparticles (Bankar, Joshi, Kumar, & Zinjarde, 2010; Emaga,

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Robert, Ronkart, Wathelet, & Paquot, 2007). It was found that the silver particles in nano-scale exhibit high antibacterial activity and have no insufferable cytotoxic effects for human beings (Damm, Münstedt, & Rsch, 2007; Damm, Münstedt, & Rsch, 2008; Kumar & Munstedt, 2005; Li et al., 2008; Liau et al., 1997; Morones et al., 2005). The antibacterial effect of silver nanoparticles has been tested for yeast and *E. coli* by Sondi and Salopek-Sondi (2004). In this study bio-inspired silver nanoparticles were synthesized with the aid of a novel, non-toxic, lignocellulosic material namely rice straw powder. Our target in this study was to develop a novel silver-based antibacterial materials that durable and compatible with the currently technologies. Here we provide economical method for coating recycled carton by polymer nanocomposites aiming to improve its mechanical and barrier properties.

2. Materials and method

2.1. Materials

Rice straw (RS) was obtained as by-product of rice crop. Commercial polystyrene (PS) was obtained from CHI MEI Corporation, Taiwan (*Mw* 97 × 10^3 g/mol, Mn 2.7 × 10^3 g/mol, D 35). Silver nitrate (Min assay 99.8%) was obtained from S.d.fiNE-CHEM LTD. Co. Toluene was obtained from El Nasr Pharmaceutical chemicals Co. Unless otherwise noted, all other chemicals were reagent grade and used as received.

2.2. Experimental part

2.2.1. Preparation of recycled carton sheet

In this work newsprint was used as waste paper. Waste paper was cut into small pieces and immersed in warm sodium hydroxide solution (5% based on oven dry weight of waste paper) at 50 °C for 2 h, then mechanically stirred. The obtained pulp was washed with water till neutrality, filtered and beaten in a valley better for 8 min to obtain slurry. Carton sheets of basis weight of 150 g/m^2 were made from this slurry according to Tappi-Standard Method using the sheet former of AB Lorentzen (Stockholm, Sweden). The sheets were then placed for conditioning at 65% relative humidity and temperature ranging from 18 to 20 °C.

2.2.2. Preparation of Ag nanoparticles and coating solution

For all experiments, the source of silver was silver nitrate ($AgNO_3$) in distilled water, where 2 g of rice straw powder was immersed in 200 ml of 5 mM silver nitrate solution. The mixture was incubated in a water bath for 3 min at 80 °C and stirred for ½ h. The reaction mixtures were observed for color change at different time intervals. The reaction mixture solution was centrifuged at 5000 rpm and the clear supernatant was second centrifuged at 13,000 rpm for Ag nanoparticles precipitation. The precipitate was dried in oven at 70 °C. Then the prepared silver nanoparticles were added to solution of polystyrene dissolved in toluene solvent into different ratios (2, 4, 6 and 8%) based on the polystyrene concentrations.

2.2.3. Coating of recycled carton sheet by PS/Ag nanocomposites

The prepared sheet was coated by polystyrene/silver (PS/Ag) nanocomposites solution with different ratios of silver nanoparticles (0, 2, 4, 6, 8%). The sheet was immersed into the solution for 1 min and air dried at room temperature. In otherwise, the silver nanoparticles were added to the pulp during making paper sheet and previous to impregnation in polystyrene solution sample no. V. The sheets were tested for tensile strength according to German Standard method by means of a Karl Frank 468 tester (Weinheim–Berkenau) and burst strength according to TAPPI Standard test method403.A.Mullen (Perkins, Chicopee, MA, USA) was used.

2.2.4. The antimicrobial study of the prepared sheet

The disc diffusion method was used to determine the antimicrobial activity of the prepared coating recycled carton by polymer nanocomposites. A volume of 0.1 ml of the tested microorganisms grown in Brian Heart Infusion Broth (at 42 °C for 24 h, 108–109 cells/ml), was inoculated on Brian Heart Infusion media, and then spread on the entire surface of the dish using a sterile spatula. Subsequently, sterile discs were placed onto agar at certain intervals by passing gently. After the plates were incubated at 42 °C for 24 h, the inhibition zones around the discs where no growth occurred, were measured in millimeters, the experiments were repeated in duplicated for all of the test strains.

3. Characterization

The nanosturacture of sliver was elucidated by JEOL JEM-1230 transmission electron microscope (TEM) with acceleration voltage of 80 kV. The microscopy probes of the sliver nanoparticles was prepared by adding a small drop of the water dispersions onto a Lacey carbon film-coated copper grid then allowing them to dry in air. Also, scanning electron micrographs (SEM) were taken using FEI INSPECTS Company, Philips, Holland environmental scanning without coating. The XRD patterns of the silver nanoparticles and recycled carton sheet were carried out on a Diano X-ray diffractometer using CoKα radiation source energized at 45 kV and a Philips X-ray diffractometer (PW 1930 generator, PW 1820 goniometer) with CuK radiation source (λ = 0.15418 nm). The basal spacing (d_L) was calculated from the (0 0 1) reflection via the Bragg′s equation. UV-spectroscopy was carried by Shimadzu UV–vis recording spectrophotometer UV-240. Water vapor permeability of carton sheet was carried out using TSY-TI equipment.

4. Result and discussion

Packaging materials offer physical protection and generate appropriate physicochemical conditions for goods that are essential for obtaining a satisfactory shelf life. The packaging system, based on a proper alternative of the packaging materials artistic with appropriate gas and water vapor barrier and mechanical properties, prevents product deterioration attributable to physicochemical or biological factors and maintains the overall quality during storage and handling. Furthermore, the using of packaging materials, such as shopping bags, still easily perceptible in the environment in many countries. So recycled carton coated by PS/Ag nanocomposites gives a possible alternatives to the traditional non-biodegradable polymers and other packaging tools, especially in short life term applications also it is very economical procedure. In the current work, we reveal the preparation of green biological route for the synthesis of silver nanoparticles via an extract resulting from rice straw powder. The prepared silver nanoparticles have been characterized by UV–vis spectroscopy, SEM, and XRD analysis. In addition, silver nanoparticles were added to the polystyrene solution by different ratios, then recycled carton sheet impregnated into the solution of PS/Ag nanocomposites, and study the mechanical and antibacterial properties.

4.1. UV–vis spectroscopy and visual observations

The reaction mixtures turned yellow-brown after (½ h, and 3 h) of incubation of sliver nitrate in presence of rice straw which can reduce silver ions to silver metal in form of nanoparticles as revealed by TEM images. The color and the intensity of the peaks

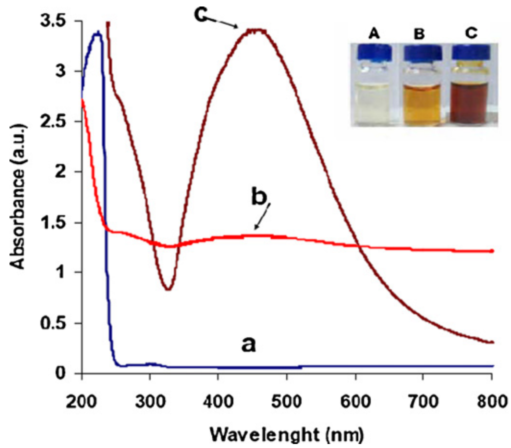

Fig. 1. Visual observations and UV–vis absorption spectra of (a) silver nitrate solution, (b) reaction mixtures containing 2 g of RS powder incubated at 80 °C with 5.0 mM silver nitrate stirring for ½ h, (c) reaction mixtures containing 2 g of RS powder incubated at 80 °C with 5.0 mM silver nitrate stirring for 3 h.

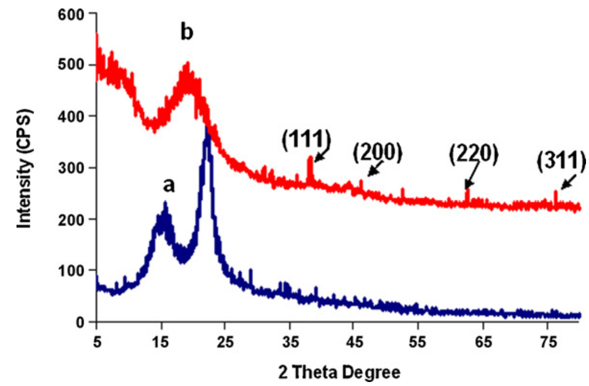

Fig. 2. XRD profiles of (a) uncoated carton sheet, (b) carton sheet coated by PS/Ag nanocomposites with 8% silver nanoparticles.

were time dependent. When using silver nitrate without rice straw, there was neither a change in color nor a characteristic peak of the solution of the same concentration of silver nitrate (Fig. 1, tube A and curve a). Control silver nitrate solutions (without rice straw) neither developed the brown colors nor did they display the characteristic peaks. These results indicated that abiotic reduction of silver nitrate did not arise under the reaction conditions. A yellow color was observed when the reaction take place using reaction mixtures containing 2 g of RS powder incubated at 80 °C with 5.0 mM silver nitrate at reaction times 3 min (Fig. 1, tube B and curve b), and turned to a dark brown color and an intense peak were observed after 3 h (Fig. 1, tube C and curve c). A change in color was also associated with well-defined peaks characterized by maximum centered around 440 nm (Fig. 1). Such peaks are recognized to be due to the plasmon resonance demonstrated by silver nanoparticles (Mulvaney, 1996).

4.2. X-ray diffraction (XRD)

The XRD profile of recycled carton sheet (not reacted with silver nitrate) is shown in Fig. 2a. The pristine recycled carton sheet without silver nitrate solution demonstrate the characteristic peaks at $2\theta = 15.9$ and 22.9 Å which related to the cellulosic fibers. After the reaction of RS with the silver nitrate and added to polystyrene solution with 8% silver nanoparticles, the X-ray diffraction pattern that was obtained and shown in Fig. 2b reveals that the characteristic peaks of pure recycled carton sheet were broadening or vanish as a result of coating with PS/Ag nanocomposites. In addition, the diffraction peaks at $2\theta = 38.1^\circ$, 46.2°, 62.64° and 76.3° allocated to the (1 1 1), (2 0 0), (2 2 0) and (3 1 1) planes of a faced center cubic (fcc) lattice of silver nanoparticles were obtained. The results of X-ray diffraction patterns showed here are matching with previous work by Satishkumar et al. (2009) and Bar et al. (2009).

4.3. Morphological study of the prepared sheet

The achievement of the formation of Ag nanoparticles step was examined based on the obtained morphological data using transmission electron microscope (TEM). Fig. 3a and b shows the formation of silver nanoparticles when a mixture of 2 g rice straw and 5.0 mM silver nitrate solution was kept under stirring for three hours at 80 °C. Furthermore, the particles size of the prepared silver nanoparticles in the range 9–30 nm.

The study showed that SEM results when appropriately interpreted and combined with TEM results offer a much clearer of formation of Ag nanoparticles. Complementary, these two techniques give information to help us to obtain significant relationships between the PS/Ag nanocomposites and recycled carton sheet. Fig. 4a and b represents SEM micrographs of the pure recycled carton sheet which maintain the homogeneity and uniformity of the surface of the prepared paper sheet. While the coated recycled carton sheet using PS/Ag nanocomposites fashioned high homogeneity of the dispersed Ag nanoparticles on the polystyrene film that covered the surface of the paper sheet the white arrow refers to the silver nanoparticles doped into polystyrene matrix. A comparison of the original sheet matrix with

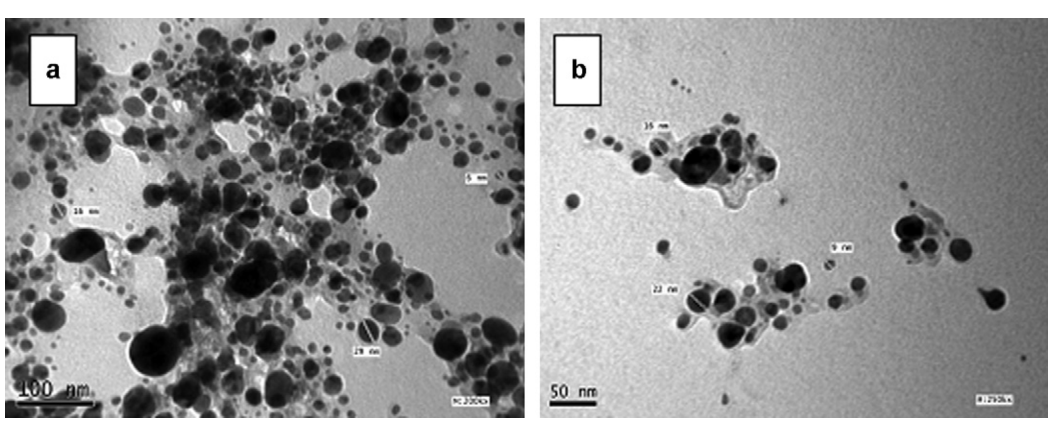

Fig. 3. Transmission electron micrographs of silver nanoparticles synthesized with 0.5 g of RS powder, in presence of 5.0 mM silver nitrate incubated at 80 °C for ½ h (a and b).

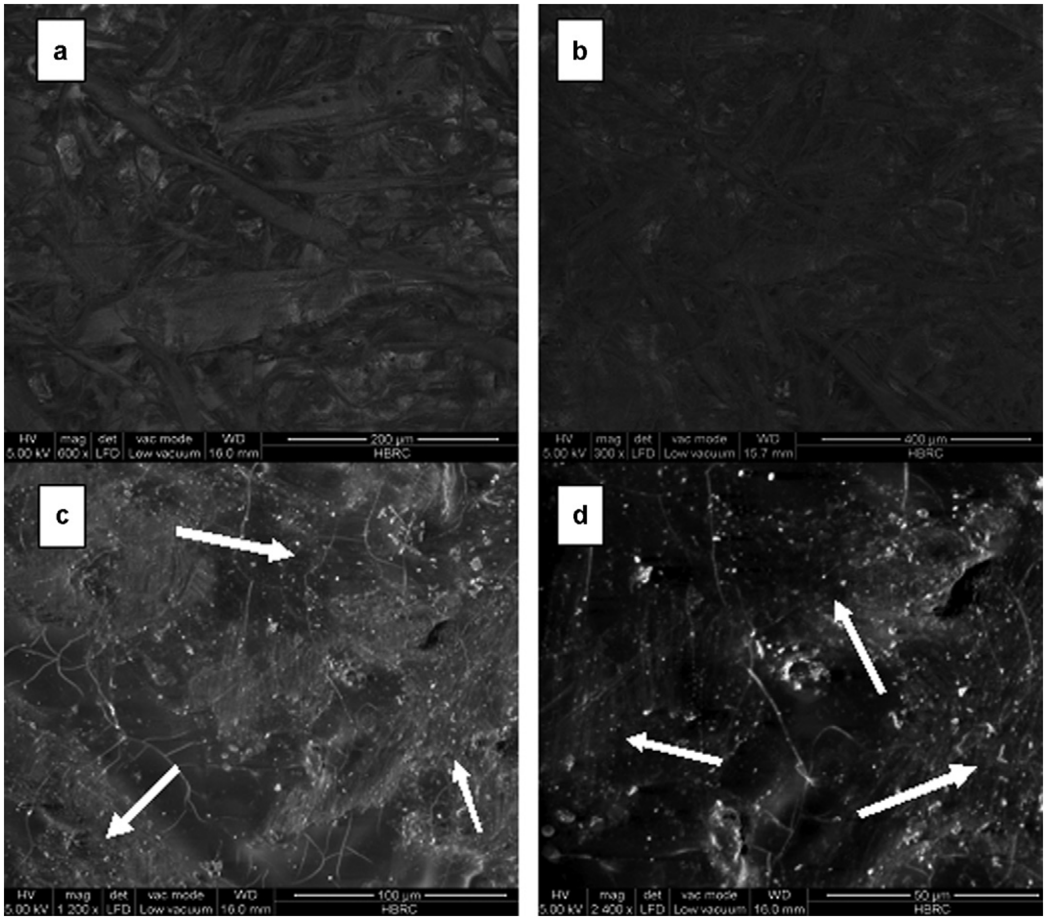

Fig. 4. Scanning electron micrographs of uncoated carton sheet (a and b), as well as the carton sheet coated by PS/Ag (8%) nanocomposites (c and d); white arrow refers to Ag nanoparticles.

that of the PS/Ag-cellulosic waste raw materials nanocomposites reveals that although the polymer appear to form largely on the surface of recycled carton sheet, the open structure of the cellulosic matrix has been retained. This offers a large accessible surface area of the polymer for chemical and physical interactions and antibacterial properties.

4.4. Mechanical and physical properties of the prepared carton sheet

The demand for recycled fiber is globally increasing by more than 4% annually compared to around 2% for virgin fibers. This means that more paper needs to be recovered and recycled. At the same time higher quality of the final deinked pulp is demanded. In this study newsprint was deinked using 5% sodium hydroxide solution based on raw material. Sodium hydroxide (NaOH) is added for reasons such as ink detachment and ink dispersion. It is commonly believed that ink detachment is improved using sodium hydroxide both because of fiber swelling effects as well as chemical hydrolysis of some bonds between the substrate and some ink species.

High tensile strengths are generally necessary for packaging carton in order to withstand the normal stress encountered during their application, subsequent shipping, and food handling. In this study tensile strength of control carton was 2360.41 m. This value was changed by dipping in polystyrene solution and more improved when silver nanoparticles were incorporated into poly styrene solution up to (8% based on polystyrene). When silver nanoparticles (2% based on polystyrene) was first added to the pulp during making paper sheet before coating by polystyrene, the tensile strength was reduced to 2199 m. This may be due to nanoparticles fill pores of paper preventing good absorption of polystyrene solution which play a role in improving tensile strength of recycled carton. Table 1 indicates that burst strength of coated carton was enhanced compared to uncoated one 2.45 kg/cm^2. In general, water vapor permeability (WVP) of carton is expected to decrease with coating by hydrophobicity of hydrocarbon which regulates the water vapor transmission rate through carton fibers. Also, Table 1 indicates that the incorporation of silver nanoparticles leads to easier water vapor migration through carton sheet fibers.

4.5. Antibacterial activity of the silver nanoparticles

Silver nanoparticles displayed antimicrobial activity towards the tested pathogenic strains of *Pesudomonas*, *S. aureus*, *E. coli*, *Salmonella*, and *B. cereus*. Waste raw materials itself did no exhibit antimicrobial activity. In all the figures, the black arrows indicate the wells of the samples and the red arrows represent the inhibition zones. In a similar behavior, antibacterial activity was observed towards *Pesudomonas*, *S. aureus*, *E. coli*, *Salmonella*, and *B. cereus*.

In Fig. 5 the antibacterial activity of *Pesudomonas* did not display any characteristic zones of inhibition which indicting that this culture were not inhibited by silver nanoparticles. The result obtained from biological method with *Pesudomonas*, *S. aureus*, *E. coli*, *Salmonella*, and *B. cereus* strains were revealed in Fig. 5. These cultures were inhibited by the bio-inspired silver nanoparticles and an inhibition zone with an average size 15–0.9 mm as shown in Table 2 and the effect of silver nanoparticles appear to be not have higher effect, this may be as a result of that polystyrene

Table 1
Mechanical and physical properties of polystyrene coated carton sheet.

Samples	Breaking length (m)	Burst (kg/cm^2)	Water vapor permeability (g/m^2)
Control	2360.41	2.45	364.23
I	2664.06	3.15	95
II	2403.2	4.05	98.51
III	2801	3.35	120.40
IV	2828.5	3.88	173.62
V	2199	3.98	157.58

I, carton coated by polystyrene; II, carton coated by polystyrene nanocomposites (2% nano); III, carton coated by polystyrene nanocomposites (4% nano); IV, carton coated by polystyrene nanocomposites (8% nano); V, carton nanocomposites (2% nano) coated by polystyrene solution.

Table 2
The antibacterial effect of carton sheet coated with PS/Ag nanocomposites (zone of inhibition in mm).

Samples	Strains				
	Pesudomonas	*S. aureus*	*E. coli*	*Salmonella*	*B. cereus*
Blank	Nil	Nil	Nil	Nil	Nil
2%	Nil	8 mm	12 mm	3 mm	7 mm
4%	Nil	0.9 mm	15 mm	8 mm	5 mm
6%	Nil	2 mm	5 mm	7 mm	4 mm
8%	Nil	12 mm	15 mm	1.5 mm	17 mm

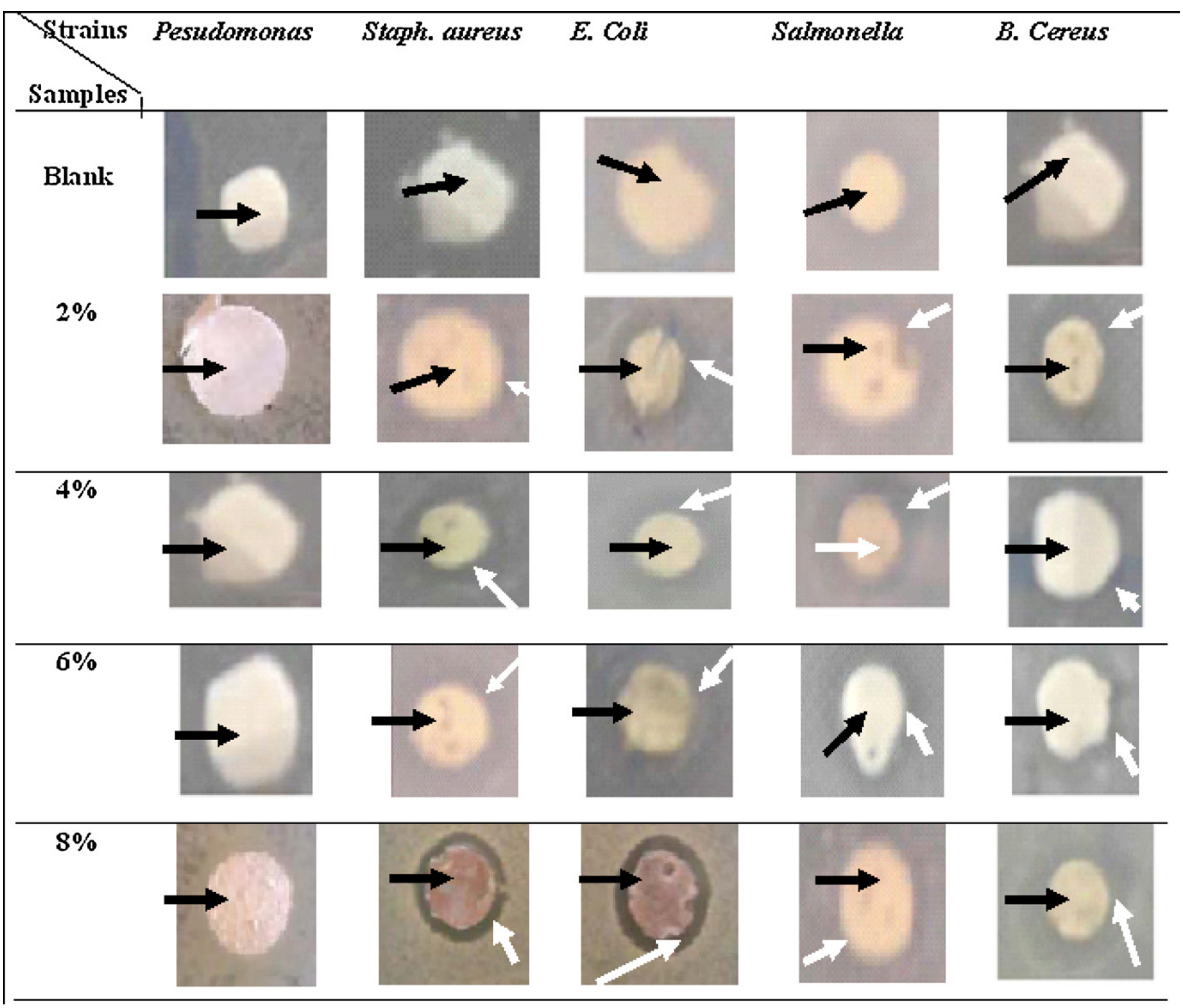

Fig. 5. The antibacterial effect of the carton sheet coated by PS/Ag nanocomposites containing different silver nanoparticles on different strains.

film making shielding effect consequently decrease the effect of silver nanoparticles and this method was reported by (Ehre, Mamane, Belenkova, & Markovich, 2009). Moreover, the achievable antibacterial activities demonstrated by silver nanoparticles have made them promising applicants as new cohort antimicrobials (Rai, Yadav, & Gade, 2009; Smetana, Klabunde, Marchin, & Sorensen, 2008). Additionally, these bio-inspired nanoparticles may also be used as additives in water paints or cotton fabrics (Khaydarov et al., 2009) on such nanoparticles illustrating marked antimicrobial effects.

5. Conclusion

Silver nanoparticles successfully prepared by novel method as antimicrobial additive, the synthetic process takes place with aid of a novel, non-toxic, and eco-friendly biological materials namely rice straw powder. The prepared silver nanoparticles were impregnated into commercial polystyrene. The recycled carton sheet was coated by PS/Ag nanocomposites by different ratios (2, 4, 6 and 8%) based on polystyrene. The X-ray diffraction pattern (XRD) and transmission electron microscope (TEM) demonstrated the formation of silver nanoparticles. The recycled carton sheet coated by PS/Ag nanocomposites exhibited antibacterial activity towards different bacterial strains. Also, the mechanical properties of recycled carton sheet coated by PS/Ag nanocomposites were increased by the coating process. Furthermore, the water vapor permeability was improved by the coating process.

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