

Effect of graphene oxide nanoparticle coatings on the strength of packaging paper and its barrier and antibacterial properties

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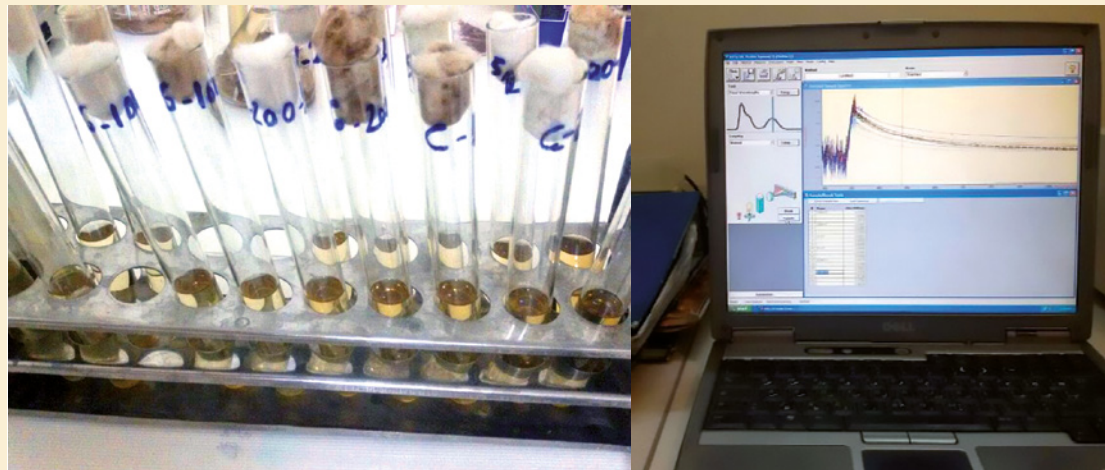


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RÉSUMÉ

Effet des couches de nanoparticules d'oxyde de graphène dans la résistance du papier d'emballage, ses propriétés de barrière et son activité antibactérienne

L'objectif de cette étude était d'évaluer la performance des nanoparticules d'oxyde de graphène dans des formulations de papier d'emballage pour améliorer les propriétés antibactériennes, physiques et mécaniques du carton. Le papier était recouvert avec des nanoparticules d'oxyde de graphène de concentrations de 100 et 200 ppm enduites avec 5 % d'amidon cationique (poids sec) comme aide à la rétention et pour un positionnement plus homogène des particules d'oxyde de nanographène sur la surface du papier. Les surfaces du papier enduites de particules d'oxyde de nanographène ont été caractérisées à l'aide des méthodes ATR-FTIR et SEM. Le test antibactérien a été réalisé selon la méthode de la turbidité. Pour les tests antibactériens des feuilles de papier, *Escherichia coli* et *Staphylococcus aureus* ont été utilisées comme bactéries à Gram-négatif et à Gram-positif, respectivement. Les résultats ont montré que l'absorption d'UV a été réduite et que la réduction la plus grande a été obtenue en utilisant des particules d'oxyde de nanographène de 200 ppm. La turbidité dans les échantillons qui incluent *S. aureus* était aussi plus basse. Le taux de croissance des bactéries *S. aureus* dans le contrôle et dans les échantillons de papier enduits d'oxyde de nanographène 200 ppm était de 89 % et de 24 %, respectivement. La densité et l'épaisseur des feuilles de papier ont augmenté dans le papier enduit d'amidon cationique et de nanoparticules, en comparaison avec le papier non enduit. Les nanoparticules n'ont pas d'effet significatif dans l'épaisseur des papiers enduits. L'ajout de particules d'oxyde de nanographène a amélioré la résistance à l'air et les propriétés de barrière des feuilles de papier. Les index de résistance à l'éclatement et la déchirure ont augmenté dans le papier enduit d'amidon et de particules d'oxyde de nanographène.

Mots-clés : oxyde de nanographène, activité antibactérienne, propriétés physiques et mécaniques.

ABSTRACT

Effect of graphene oxide nanoparticle coatings on the strength of packaging paper and its barrier and antibacterial properties

The aim of this study was to assess the performance of graphene oxide nanoparticles in paper coating formulations in order to improve the antibacterial, physical and mechanical properties of paperboard. The paper was coated with graphene oxide nanoparticles at concentrations of 100 and 200 ppm together with 5% cationic starch (dry weight) as a retention aid and for better coverage and more homogeneous positioning of nanographene oxide particles on the surface of the paper. The paper surface coated with nanographene oxide particles and starch was characterised using ATR-FTIR and SEM. The antibacterial assay was performed according to the Turbidity Method. For the antibacterial tests of paper sheets, *Escherichia coli* and *Staphylococcus aureus* were used as Gram-negative and Gram-positive bacteria respectively. The results showed that UV adsorption was reduced, with the largest reduction obtained when using nanographene oxide particles at 200 ppm. Turbidity in the samples including *S. aureus* was also lower. The growth rates of *S. aureus* bacterium in the control and the paper specimens coated with 200 ppm nanographene oxide were 89% and 24%, respectively. The density and thickness of the paper sheets increased in the paper coated with cationic starch and nanoparticles in comparison with uncoated paper. The nanoparticles had no significant effect on the thickness of coated papers. The addition of nanographene oxide particles improved the resistance to air and barrier properties of paper sheets. The burst and tear indexes increased for the paper coated with starch and nanographene oxide particles.

Keywords: nanographene oxide, antibacterial activity, physical and mechanical properties.

RESUMEN

Efecto de los recubrimientos de nanopartículas de óxido de grafeno en la resistencia, las propiedades de barrera y la actividad antibacteriana del papel de envolver

El objetivo de este estudio fue evaluar el rendimiento de las nanopartículas de óxido de grafeno en formulaciones de recubrimiento de papel para mejorar las propiedades antibacterianas, físicas y mecánicas del cartoncillo. El papel se recubrió con nanopartículas de óxido de grafeno con concentraciones de 100 y 200 ppm acompañadas de almidón catiónico al 5% (porcentaje respecto al peso en seco) como ayuda a la retención, mejor cobertura y posicionamiento más homogéneo de las partículas de óxido de nanografeno en la superficie del papel. La superficie del papel recubierta con partículas de óxido de nanografeno y almidón se caracterizó mediante ATR-FTIR y SEM. La prueba antibacteriana se realizó según el método de la turbidez. Para las pruebas antibacterianas de las hojas de papel se utilizaron también *Escherichia coli* y *Staphylococcus aureus* como bacterias gram-negativa y gram-positiva, respectivamente. Los resultados mostraron que la absorción de UV se redujo, y que la mayor reducción se obtuvo al utilizar partículas de óxido de nanografeno a 200 ppm. También fue menor la turbidez en muestras que incluyen *S. aureus*. La tasa de crecimiento de *S. aureus* bacterium en el control y en muestras de papel recubierto con óxido de nanografeno de 200 ppm fueron del 89% y 24%, respectivamente. La densidad y el espesor de las hojas de papel aumentó en el papel recubierto con almidón catiónico y nanopartículas en comparación con el papel no recubierto. Las nanopartículas no tienen efecto significativo en el grosor de los papeles recubiertos. La adición de partículas de óxido de nanografeno mejoró la resistencia al aire y las propiedades de barrera de las hojas de papel. Los índices de resistencia a la explosión y al desgarro aumentaron en el papel recubierto con almidón y partículas de óxido de nanografeno.

Palabras clave: óxido de nanografeno, actividad antibacteriana, propiedades físicas y mecánicas.

Introduction

Paper substrates are widely used in many industries such as packaging due to their high strength, flexibility, low cost and recyclability. However, they have a complex structure, with cellulosic units arranged at multiple levels including molecules, fibrils, fibers and fiber networks (Afra *et al.*, 2013). Paper and its products are closely related to our lives and work. Because paper contaminated by bacteria gives rise to the spread of disease, increasing attention has been focused on the research of antibacterial paper (Deng *et al.*, 2012).

With the growing public health awareness of disease transmissions and cross-infection caused by microorganisms, use of antimicrobial materials has increased in many application areas like protective clothing for medical and chemical works, other health related products (Duran *et al.*, 2007), antibacterial packaging material that can improve product quality and keep it free from microbial adhesion (Park and Zhao, 2004), etc. It is of great significance to study antibacterial paper in order to prevent the spread of harmful bacteria and infection of infectious disease. The antibacterial paper is putting a small amount of antimicrobial agent to spraying, dipping, sizing, coating, the modified fiber added to ordinary paper. The antibacterial agent generally can be divided into inorganic antimicrobial agent, organic antibacterial agent and natural antimicrobial agent (Hong and Tang, 2014). The advantages of coated paper is low cost, wide source, and easy to recycle. It will not cause pollution to the environment, complying with the principles of green packaging. Coating ingredient that has a crucial influence on the application of product is the most basic technology; manufacturing coated paper has a crucial influence on product use (Pang *et al.*, 2015). Nanotechnology is presently recognized as one of the most promising areas for packaging technological improvement in the 21st century (Hu and Fu, 2003). Nanoparticles usually ranging in dimension from 1-100 nanometers (nm) have properties unique from their bulk equivalent.

Moreover, nanoparticles possess increased surface area and therefore increase the area of interaction with the pathogenic bacteria. They are also more likely to enter the bacterial surfaces than micron particles due to smaller size, exerting stronger effects on bacterial targets (Tran and Webster, 2011). Improving results were also reported against exposure to some fungi and insects by addition of metal and mineral nanoparticles (Taghiyari *et al.*, 2019; Akhtari and Nicholas, 2013). But one major problem for metal-based nanoparticles is their toxicity to healthy cells due to the generation of reactive oxygen species (Xia *et al.*, 2008). Those materials may result in severe health problems when such coated paper products are used for food wrapping or clinical applications.

Graphene Oxide (GO) is chemically modified graphene,

containing hydroxyl, carbonyl and epoxy functional groups, which is obtained by synthesis of graphite with strong oxidizing agents (Alves *et al.*, 2014). Also it has been used as a promising material for preparing new composites (Tang *et al.*, 2013). It is well known that GO and its composites possess antimicrobial properties and have been used as antibacterial and antifungal agents (Santos *et al.*, 2012; De Faria *et al.*, 2014). Nanotechnology and nanomaterials have been effective to improve properties of many materials and overcome some of their drawbacks (Majidi, 2016). In the present study, we evaluated the performance of graphene oxide nanoparticles in the paper coating formulations in order to improve the antibacterial, physical, mechanical and mechanical properties of paper.

Materials and methods

Materials

Nanographene oxide particles with thickness of 3.4-7 nm, average number of layers 6-10, surface area of 100-300 m²/g, ~99% carbon purity and bulk density of 1 g/cc were purchased from Nanosany Co., Ltd. Figure 1 shows a microphotograph of the layer structure nanographene oxide. Cationic starch was obtained from Merck Chemicals Co., Ltd. A4-size paper sheets with a grammage of 80 g/m² and an average thickness of 112 µm with trade name Copimax was use as a controlled paperboard for the coating.

Preparation of coating solutions

In this study, the paper was coated with graphene oxide nanoparticles with concentrations of 100 and 200 ppm accompanied by cationic starch at the 5% percent (based on dry weight) as a retention aid, better coverage and more homogeneous positioning of nanographene oxide particles on the paper surface. The starch coating solution by gelatinizing 5% concentration was prepared by adding 5 g starch in the 100 mL distilled water. This mixture cooked at a temperature of 90 °C for 30 minutes. It was kept in a constant temperature water bath at 50 °C and used it for surface coating. Graphene oxide nanoparticles suspension 100 ppm and 200 ppm were prepared by adding respectively 100 mL and 200 mL colloids solutions of graphene oxide nanopar-

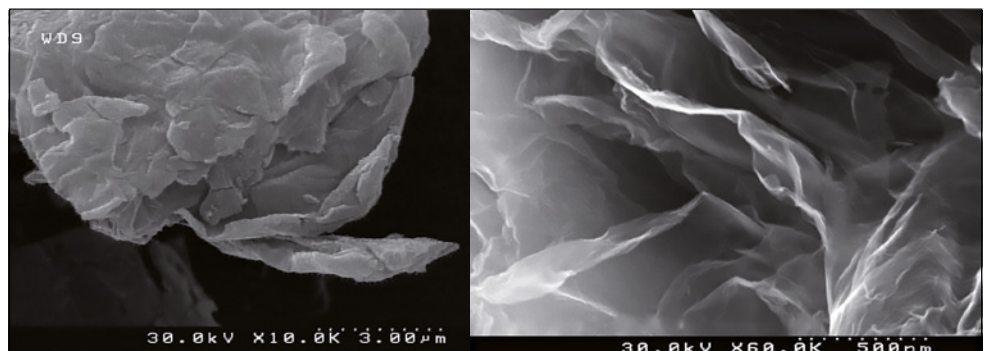


Figure 1.
SEM image of graphene oxide nanoparticles as the layer structure.

ticles in the 900 mL and 800 mL gelatinize starch solution. The mixtures were then homogenized and stirred using a magnetic stirrer unit for 30 min.

Paper sheets coating

Coating solutions were coated onto paper sheets using the Auto Bar Coater (GBC – A4 GIST Co., Ltd). 10 mL of coating solutions was poured on the paper along the width at one end and the applicator rod was immediately swept along the length of the paper sheets. The applicator speed of the coating was set at 25 mm/s. The coated paper sheets were dried at room temperature for 24 hours. Before characterization, all samples were conditioned at 27 °C with 65% relative humidity for at least 24 h in accordance with ISO 187.

Assessment of paper sheets for antibacterial properties

The antibacterial assay was performed according to the Turbidity Method. Culture medium (BHIB) was placed in an autoclave (15 min at 121 °C). Coated and uncoated sheets were sterilized in an oven (0.5 g, 2 h and 150 °C) were sterilized. Sterilized papers were inserted into a tube containing sterilized culture; then 0.05 mL with 0.05% concentration of gram negative bacteria (*Escherichia coli*) and gram positive bacteria (*Staphylococcus aureus*) were added separately

and put into a shaker incubator (160 rpm and 37 °C) for 24 h. Controls were also run in parallel. The turbidity of the suspensions was measured at 600 nm using a spectrophotometer.

Characterization of paper sheets

The surface chemical groups of the samples were measured by Attenuated Total Reflection-Fourier Transform Infrared spectroscopy (Golden Gate Model, Specac Company, UK). The surface morphology of the samples was observed by scanning electron microscopy (Vega3-Tescan, Tescan Company, Czech). The samples for SEM observations were dried at room temperature, and sputter-coated with gold before observation. All paper properties were measured according to TAPPI standards. Base Weight, Thickness, Air permeability, Water absorbion, Water Vapor Transmission Rate (WVTR), Burst strength and Tear resistance were measured according to T410om-02, T411om-05, T460om-02, T441-om-96, T448-om-09, T403om-02 and T414om-04 standards, respectively. Five specimens were selected for the evaluation of each test. Statistical analysis was conducted using SPSS software program, version 25. The Duncan test was used to compare averages and analysis of variance (ANOVA) was performed on the data to conclude significant differences at the 99% level of confidence.

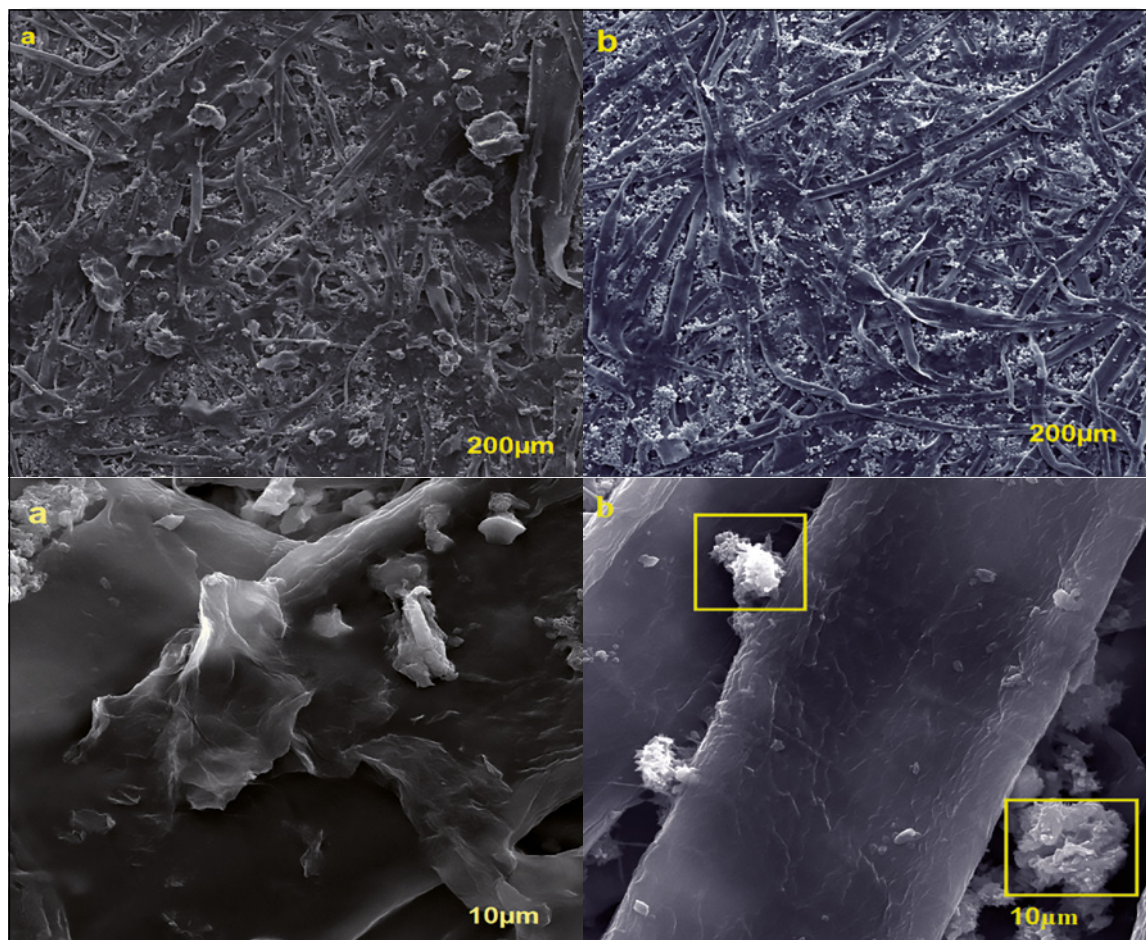


Figure 2. SEM microphotographs of the paper surface coated with graphene oxide nanoparticles (a) and starch (b).

Results and discussion

SEM and ATR characterization

SEM was employed to observe the surface morphological changes for paper samples after the particles deposition, and the representative SEM images of the coated paper with nanographene oxide particles and starch are provided in figure 2. On the nanographene oxide particles coated paper samples, the graphene oxide nanoparticles were distributed and covered the dense network of entwined cellulose fibers. Some of the graphene oxide nanoparticles were also observed in the fiber structure on the top surface. The lateral dimension for most of the nanographene oxide particles were around 10-50 μm . SEM photos of GO shows that, the exfoliated particles were smooth with small wrinkles at the edges (Al-Thani *et al.*, 2014). After the deposition of nanographene oxide particles and starch on the uncoated paper, microporous pores were filled with them and some gelatinous substances appeared and resulted in bridging between adjacent cellulose fibers. The average diameters of the cellulose fibers increased and more gelatinous substances were presented, hence causing more bridging and bonding between fibers, resulting in a more compact surface structure. This result is consistent with a previous study (Wu and Farnood, 2014; Li *et al.*, 2018).

To confirm further the coating of graphene oxide nanoparticles on the paper surface, attenuated total reflection (ATR) infrared spectroscopy were recorded. The composition of paper in general is very complex. However, the major composition of paper includes cellulose, CaCO_3 , silicates, thickeners, and binders (Ghule *et al.*, 2006). Figure 3 shows the ATR-FTIR spectra of the uncoated and coated paper with starch and nanographene oxide particles. The ATR-FTIR spectra of the papers showed absorption peaks of fibers, of which the main ingredient was cellulose. Starch has a similar sugar unit structure with cellulose and its structure is very close to that of cellulose (figure 3a, b).

Both of the ATR-FTIR spectra showed a peak in the range 3,400 to 3,200 cm^{-1} was attributed to the intermolecular hydrogen bonds. A broad band (3,100-3,700 cm^{-1}) centered around 3,336 cm^{-1} characteristic of O–H functional group (free and H-bonded) is also observed. The band at 3,336 cm^{-1} representative of free O–H shows decrease in intensity, suggesting that the hydroxyl functional groups are occupied with the deposition of starch and nanographene oxide particles (figure 3a, b).

The absorption peaks at 3,300 cm^{-1} and range from 900-1,300 cm^{-1} , respectively, can be attributed to the C–H stretching vibrations and C–O of cellulose. The band at 1,640 cm^{-1} can be attributed to the absorbed water in the cellulose fibers (Liu *et al.*, 2005). The relatively broad peak at 3,300 cm^{-1} is due to the adsorbed

water content in the surface of nanographene oxide and peak at 1,600 cm^{-1} can be assigned to the C–C vibrations from the graphitic domains (Krishnamoorthy, 2011, 2012). The FTIR spectrum of nanographene oxide in coated paper illustrates the presence of C=O stretching in carboxylic acid at 1,700 cm^{-1} , C–OH at 1,415 cm^{-1} , C–O at 1,027 cm^{-1} and C–O–C at 1,250 cm^{-1} (Zhou *et al.*, 2009).

The band at 1,735 cm^{-1} is characteristic of hemicelluloses. The weakening of absorption intensity of O–H group in peaks 1,415 cm^{-1} and 1,407 cm^{-1} indicated the decrease in free O–H group after linking with nanographene oxide particles and starch (figure 3a,b). A weak broad band at 709 cm^{-1} might be characteristic of I_α and I_β phases of cellulose. The bands at 871, 1,025, 1,159 cm^{-1} are attributed to cellulose (paper composition) (Ghule *et al.*, 2006).

Physical and mechanical properties of the paper sheets

Density, coating weight and paper thickness

Density is among the important structural properties of paper (Levlin and Söderhjelm, 1999). The apparent density of paper is also one of the physical properties of paper that impacts its strength properties. It influences almost all the mechanical, physical and electrical properties. Various hydrophilic polyelectrolytes, including cationic starch products, are used by papermakers to promote inter-fiber bonding and increase paper's dry-strength (Hubbe, 2006). The influences of nanographene oxide particles and starch on the paper's density, coating weight and thickness were also

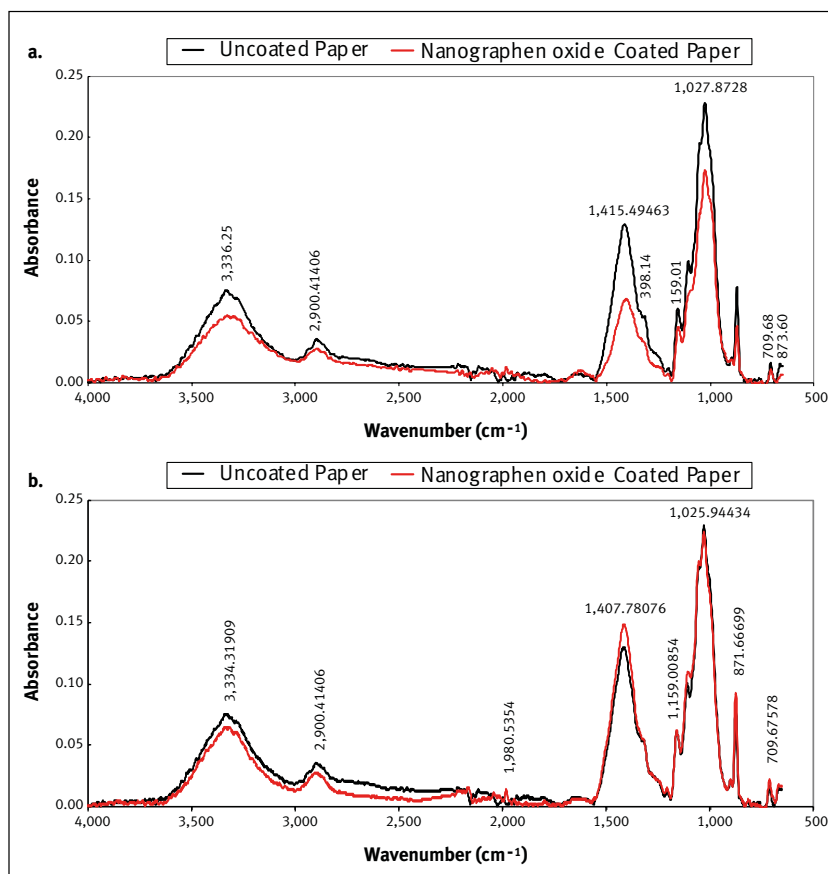


Figure 3. ATR-FTIR spectra of the uncoated and coated paper sheets with nanographene oxide particles (a) and starch (b).

evaluated: the results appear in figure 4. As it can be seen, density and coating weight of the paper sheets increased in the paper coated with cationic starch and nanoparticles in comparison with uncoated paper. Nanoparticles have no significant effect on the thickness of coated papers, which could be attributed to penetration of nanoparticles in the porous paper and low weight of nanoparticles. One-way analysis of variance revealed that there was no significant difference between average thicknesses of papers coated with nanographene oxide particles amounts of 100 ppm and 200 ppm concentrations.

Water absorption, resistance to air and water vapor transmission rate (WVTR)

It is essential for packaging material to have excellent barrier properties. For example, food packaging paper must have good oxygen and water vapor barrier properties to make sure that the food will not become bad (Wang and Jing, 2016). Figure 5 shows the barrier properties of uncoated and coated paper. Water absorption is one of the barrier properties of papers. The wetting and adsorption mechanisms of water depend on the chemical and physical heterogeneity of porous substrates, such as textiles in general and paper in specific (Werner *et al.*, 2005). The water absorption of the coated and uncoated papers was evaluated by Cobb-test measurements

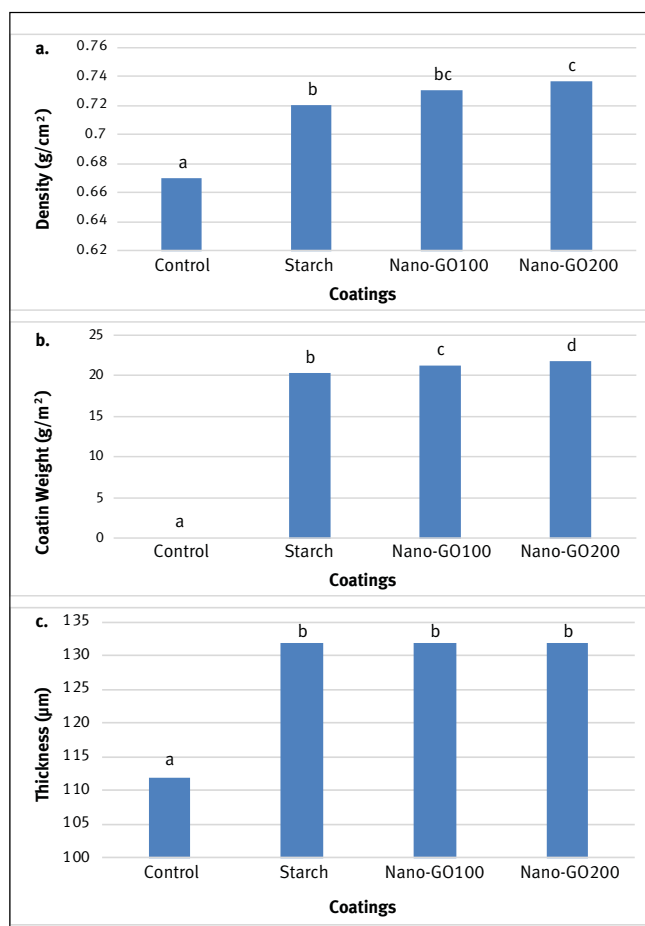


Figure 4. The effect of coatings on density (a), weight (b) and thickness (c) of the samples.

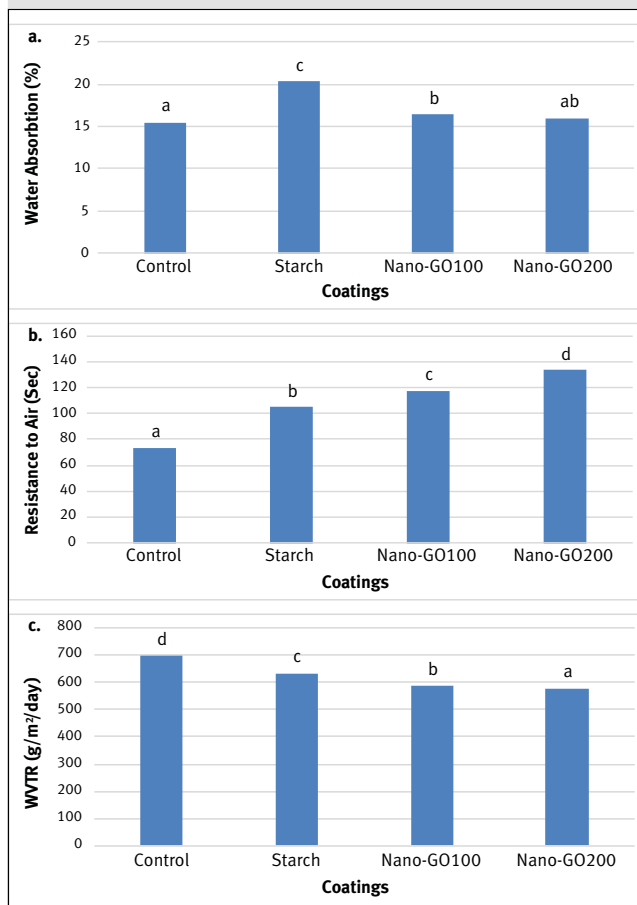


Figure 5. The effect of coatings on water absorption (a), resistance to air (b) and water vapor transmission rate (WVTR) (c) of the samples.

and displayed in figure 5a. According to the results, coated paper with nanographene oxide particles have significantly decreased. These results could be attributed to the small size of the nanoparticles and higher specific surface area. These particles are more easily placed in the pores between the fibers and reduced the amount of water absorption by filling the pores. These results were confirmed by Tunc *et al.* (2007) and Pornasir *et al.* (2016). Increasing the level of nanoparticles application reduced the absorption of sample water. This decrease has a significant effect on the water absorption properties of the samples. The increase in water absorption of coated papers with starch results from higher hydrophobicity of the paper surface. Retulainen and Nurminen (1993) showed that higher-yield fibers, having a higher density of negative charge, were able to absorb cationic starch more efficiently compared to low-yield fibers. Cationic starch is a hydrophilic polymer (Kalambur and Rizvi, 2006) with high molecular mass and cationic property, that they can induce hydrogen bond with the surface of the cellulose fibers with a negative charge (Hubbe, 2006).

Due to the hydrophilic nature of fiber surfaces, it is to be expected that hydrogen bonds, in addition to London-van der Waals (dispersion) attractions, will hold the fibers together after paper is dried (Pierce, 1930; Campbell, 1947; Davison, 1980; Fowkes, 1983).

Another important property of paper sheets is resistance to air. The objective in measurement of resistance to air of the samples was to highlight the fact that the network permeability could be better controlled in paper than in a woven fabric (Imani *et al.*, 2011). Figure 5b shows that coatings increase the resistance to air to a great extent. The highest and lowest the resistance to air values were found in coated with nanographene oxide 200 ppm (133.66 Sec) and control (73.66 Sec) paper sheet, respectively. Duncan's multi-range test puts four measurable treatments in four different groups. Addition of nanographene oxide particles improved the resistance to air and barrier properties of paper sheets. The interaction of the nanoparticle coatings with the cellulosic paper web results in improvement of the mechanical paper strength and is attributed to hydrogen-bonding between the nanoparticles and the cellulosic fibers (Samyn *et al.*, 2010).

The coating weight had a great effect on the WVTR of a coated paper. This can be seen from figure 5c, which shows that the coated paper with 200 ppm nanographene oxide particles had a low WVTR when the coating weight was high. When the coating weight was 21.70 g/m², WVTR was reduced by 577.11 g/m² day. This was because, under the equal conditions, the coated paper with higher coating weight formed a thicker layer on the surface of the paper, and these coatings were helpful to hinder water vapor from seeping through the paper. The starch and nanographene oxide particles on the paper's surface had blocked the pores on the surface from fiber and pores formed by fiber crossing; thus it had an effect on barrier function. The two factors of bond strength and bonded bonding between the fibers can be significantly increased by dry resistance additives, such as cationic starch (Mauyer, 1998). One-way variance analysis showed that there is a significant difference between measured average values at the 1% confidence level. Duncan's multi-range test puts four measurable treatments in four different groups.

Indexes of burst and tear

The mechanical properties of paper, and the strength of the fiber network, have been thought to arise from inter-fiber bonding – the strength of individual bonds and all the bonds within the network and from the (axial) strength of individual papermaking fibers (Robinson, 1980; Stratton, 1991). Tear index depends on fiber length, individual fiber strength, and Relative Bonding Area (RBA). In the case of a sheet having low RBA, tear strength is more dependent on fiber-fiber bondings, whereas by increasing of bonding, individual fiber strength becomes crucial in tear index (Testova, 2006). The influences of nanographene oxide particles and starch on indexes of burst and tear of the paper sheets were also evaluated, and the results are illustrated in figure 6.

Bursting strength tells how much pressure paper can tolerate before rupture. It is important for bag paper. Generally, bursting strength can depend on the fiber type, proportion and amount of fibers present in the sheet. For instance, an increase in basis weight and degree of refining

will result high burst strength. Fiber length and fiber coarseness have also some effect on bursting strength. Fiber length increases burst strength and coarseness decreases it (Farouk *et al.*, 1999). As can be seen in figure 6a, burst index increased in coated paper with starch and nanographene oxide particles. The burst index values in coated paper with nanographene oxide 200 ppm and control were 184.63 Pa/m²/g and 159.7 Pa/m²/g, respectively. Duncan's multi-range test puts coated papers in the same groups. The effect of coatings on the tear indexes is similar, but more slowly (figure 6b). This increase does not have a significant effect on the tear index of the samples. These results confirmed Janković-Častvan *et al.*'s report (2014), which indicated that values of the bursting and mechanical strengths of the paper coated by starch with sepiolite nanoparticles were increased.

Antibacterial activity of the paper sheets

The other important property of the paper coated with starch and nanographene oxide particles was the antibacterial activity. Here in, the Gram-negative bacteria (*Escherichia coli*) and Gram-positive bacteria (*Staphylococcus aureus*) were chosen as the test microorganism, and the antibacterial activity of the coated paper was measured using the Turbidity Method. The rate of growth of a bacterial culture is expressed as generation time, under standard nutritional conditions (culture medium, temperature, pH, etc.). Generation times for bacteria vary from about 12 min to 24 hours or more (Lindqvist, 2006). According to figure 7, for both types of bacteria *S. aureus* and *E. coli*, by increasing the amount of nanographene oxide particles, the UV adsorption was reduced and the greatest reduction was obtained when using nanographene oxide particles 200 ppm. This

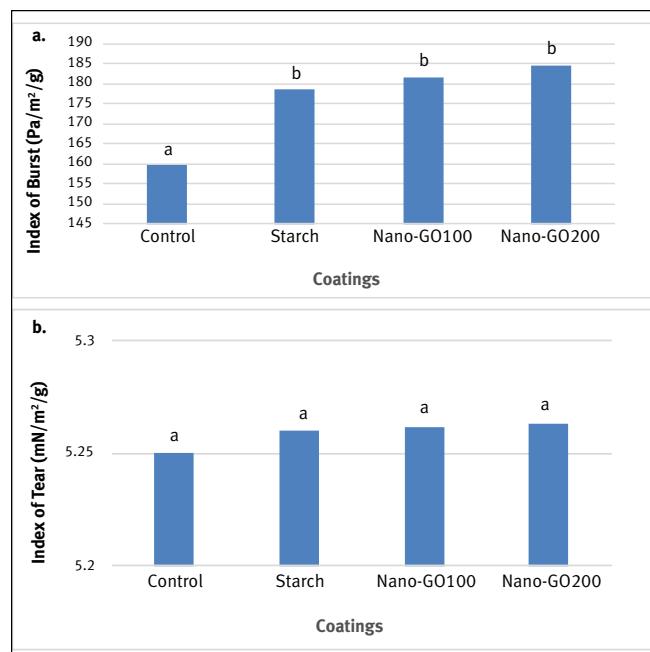


Figure 6. The effect of coatings on indexes of burst (a) and tear (b) of the samples.

shows that higher adsorption of nanoparticles led to higher anti-microbial effects in hand sheets. The results indicate that the growth rates of *S. aureus* (figure 7a) and *E. coli* (figure 7b) bacteria in the control samples were 89% and 73% respectively. With paper coated with nanographene particles 200 ppm, they reached only 24% and 34%, respectively. Coating with starch also had no inhibitory effect and no significant effect on bacterial growth on paper sheets; this result was confirmed by Pornasir *et al.* (2016). Also, turbidity in samples including *S. aureus* was lower. As a rule, Gram-positive bacteria are more sensitive to nanometal particles than Gram-negative bacteria due to a difference in the cell walls (Yazdani *et al.*, 2014). In Gram-positive bacteria, the cell wall is rich in mucopeptide component, while in Gram-negative ones the cell wall just contains a thin layer of mucopeptide and is primarily composed of lipoproteins and lipoprotein polysaccharides. Therefore, *E. coli* show more resistance to antibacterial materials (Tasou and Nychas, 1995). The present findings confirmed the other results reported by Sondi and Salopek-Sondi (2004) and Giang and Manolaches (2004). Therefore, nanographene oxide particles have a large specific surface area that enabled it to adsorb and fix bacteria effectively. It is clear in figure 7 that coated paper with nanographene particles showed higher resistance to bacteria than the control samples. Considering the efforts taken on the antibacterial activity of graphene oxide nanoparticles until now, Hu *et al.* (2010) reported the antibacterial activity of the graphene and graphene oxide towards *E. coli*. While showing minimal cytotoxicity, nanoparticles possess increased surface areas and therefore have increased interactions with biological targets (such as bacteria) compared with conventional, microparticles. Furthermore, the antibacterial effect of surface coating paper was better than that of internal additive paper, possibly because the antibacterial agent was lost in the internal additive method and was completely retained during surface coating. This is consistent with the results on physical properties (Ling *et al.*, 2013).

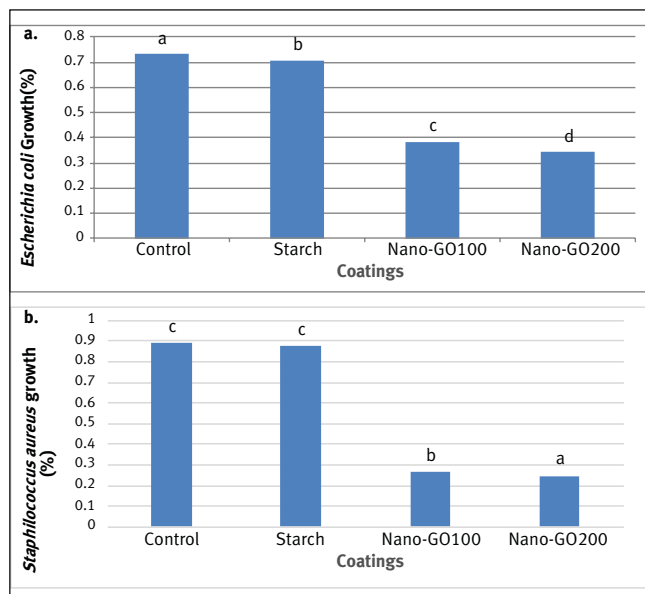


Figure 7.
Antibacterial activities of samples against *Staphylococcus aureus* (a) and *Escherichia coli* (b).

Conclusion

Antibacterial materials are widely used in daily life and effectively protect the public health. Nanographene oxide nanoparticles have been reported to demonstrate high biological activity and low toxicity. In this review, the antibacterial activity of nanographene oxide particles as a paper coating against of *Escherichia coli* and *Staphylococcus aureus* was studied by the bacterial turbidity method. Results revealed that nanographene oxide particles coatings significantly inhibited the growth of *E. coli* and *S. aureus* on the surface of papers and the antibacterial activity increased with increasing concentration. Thus, it suggests that nanographene oxide coatings could be used as an effective way.

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